BIO-ELECTROCHEMICAL CONVERSION OF CO_2 TO CH_4 TOWARDS ENRICHMENT OF BIOGAS: A REVIEW ON BES AN EMERGING TECHNOLOGY

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Abstract: The need of new sustainable sources of energy is on the top priority to fulfil the demand of energy at different corners today. Biogas is one distinct source from other renewable energy providing sources because of its environment friendly as well as economic characteristics. Biogas does not have any geographical limitations nor it requires advancedtechnology for producing energy, it is also very simple to use and apply.Although, Biogas generated from anaerobic digestion process is a clean, carbon neutral and environmental friendly energy alternative, raw biogas needs to be purified necessarily before using. The removal of impurities from raw biogas which is referred as enrichment ensures its better applications and also enhances its calorific value. Fundamental aspects of anaerobic digestion are briefly described in the paper. The paper mainly includes various biogas enrichment techniques and their comparative studies have been made. A novel technique of bio-electrochemical technique has also been briefly discussed.

Keywords: fossil fuels, biogas, bio-methane, renewable, enrichment, bio-electrochemical

I. INTRODUCTION

Biogas is produced from controlled anaerobic digestion of organic compounds such as animal waste, cellulosic biomass and food wastes. Raw biogas composed of CH_4 (60-75%), CO_2 (25-30%), and minor concentrations of other gases including H_2, NH_3, H_2S, and other. Currently, raw biogas is not only used for cooking but also used for vehicular operations and electricity generation. In India, Ministry of New and Renewable Energy had set a target to achieve 48.55 MW energy from biogas plants till 2022. Due to its low calorific value and impurities, raw biogas is not readily suitable for application as a source of energy. In order to meet the specifications of natural gas to utilized raw biogas as a vehicular fuel (known as bio-methane or bio natural gas), CH_4 content should be upgraded to at least 95%. Thus, enrichment of biogas in terms of CH_4 content required through removing CO_2 or converting CO_2 to CH_4.Upgradation of raw biogas to a higher level of methane content has already been developed and utilized as compressed biogas (CBG).

Biogas enrichment methods include physical, chemical, biological, or a combination of these approaches. Physical upgrading involves removing CO_2 from biogas through techniques such as organic solvent absorption, water scrubbing, pressure swing adsorption, cryogenic separation, or membrane separation. Chemical upgrading reduces CO_2 to CH_4 using H_2 as a reductant. Biological upgrading methods utilize photosynthetic or chemoautotrophic reactions. To enhance methane content for uses like transport fuel, numerous physical and chemical upgrading technologies have been developed and reviewed. These conventional, ex-situ methods focus on extensive CO_2 removal but have drawbacks, including high costs, energy requirements, and sometimes toxic solvents to achieve 95-99% methane content. Studies show that these techniques are not widely adopted due to their economic and energy inefficiency for smaller scales. Currently, only a few commercial plants globally use these methods to upgrade biogas to high fuel standards, and the CO_2 removed during physical cleaning is often released, contributing to global warming.

Unlike physical and chemical methods, biological technologies use microorganisms to convert CO_2 into CH_4 at moderate temperatures and atmospheric pressure. These methods are increasingly favored because they capture and recycle CO_2 into new products, resulting in smaller carbon and energy footprints. Recently, the microbial bio-electrochemical process (MBE) has been explored as a promising alternative for upgrading biogas on-site. Anaerobic microorganisms can utilize a variety of organic compounds for carbon and energy. The primary aim of an anaerobic digester is to biologically break down a significant portion of the volatile solids in sludge and reduce its putrescibility. Anaerobic digestion (AD) is a complex biochemical process where organic materials are decomposed by a consortium of microorganisms in the absence of oxygen. Biogas upgrading enhances the energy content of biogas (i.e., CH_4 concentration) to enable its use in a broader range of applications without needing a CHP unit. This can facilitate the direct use of biogas for on-site energy recovery (e.g., CH_4-powered vehicles) or allow for its injection into existing natural gas infrastructure if the CH_4 content exceeds 96% (v/v). However, there are significant challenges, and bio-methanation is now considered unsuitable for industrial applications. Ex situ biogas upgrading methods such as adsorption, absorption, membrane systems, cryogenics, and algal biomass systems are available but often involve high costs for consumables, substantial energy requirements, or large space needs.

Electrochemical reduction involves converting carbon dioxide, a low-energy component of biogas from anaerobic digesters, into methane, an energy-rich component. This process occurs through a reaction between CO_2, protons, and electrons (supplied by electricity) in a microbial electrosynthesis system (MES). This enables biogas (bio-methane) from waste treatment plants to be directly integrated into existing gas grids or used as a transport fuel. Combining anaerobic digestion with anodic oxidation from MECs has enhanced wastewater treatment. Rapid advancements in bio-electrochemical technology make it a promising method for CO_2 capture and conversion compared to other approaches. With a small voltage applied to the MEC, electromethanogens can directly convert CO_2 into CH_4.

This article covers the basics of anaerobic digestion and highlights various upgrading techniques currently under investigation. Conventional technologies for biogas enrichment are discussed, alongside the innovative bio-electrochemical systems.

II. FUNDAMENTALS OF ANAEROBIC DIGESTION

Anaerobic microorganisms can derive carbon and energy from a diverse array of organic compounds. The purpose of an anaerobic digester is to biologically decompose a significant portion of the volatile solids in sludge and reduce its putrescibility. Anaerobic digestion (AD) is a complex biochemical process where organic materials are broken down by a group of microorganisms in the absence of oxygen. Figure 1 illustrates how AD efficiently converts organic matter into biogas, with methane (CH_4) being the primary combustible component. In India, various anaerobic digestion technologies are employed, each differing in design based on factors such as feedstock solids content, process stages, operation mode, and temperature. Understanding these factors helps project implementers select the most appropriate technology to suit their specific needs.

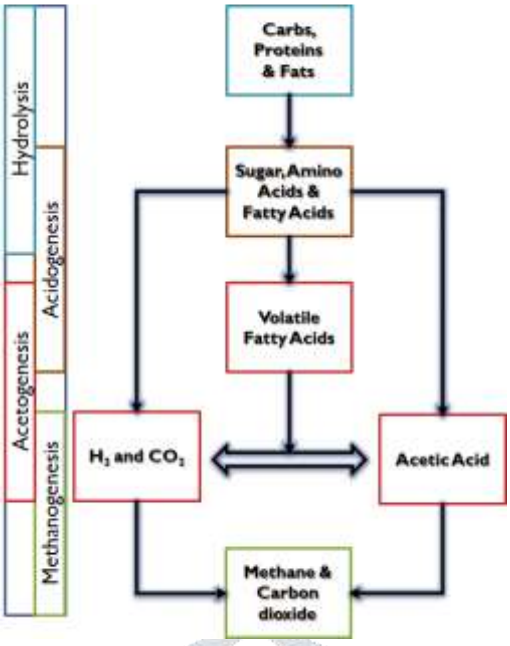

Fig. 1: Conversion of organic matter into a valuable product

The technology (AD) has a positive net energy production rate and the CH_4 gas produced from the process also has the tendency to replace fossil fuels. In fact, if properly handled, AD systems have no negative effect on human health or on the environment. Degradable complex organics go through four stages during anaerobic digestion as shown in Figure 2: hydrolysis, acidogenesis, acetogenesis and methanogenesis.

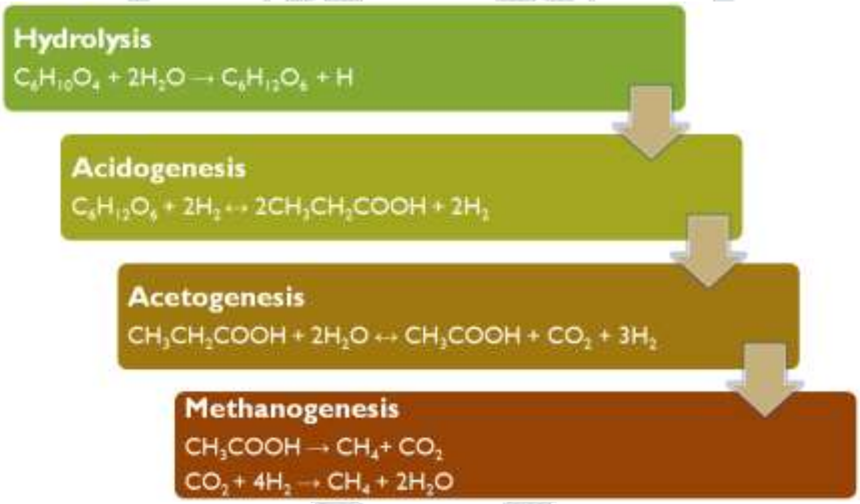

Fig. 2: Steps of Anaerobic Digestion Process

The resulting biogas, produced from anaerobic digestion, consists of approximately 30-40% carbon dioxide (CO_2) and 60-70% CH_4 along with other trace gases (e.g., H_2S, N_2, H_2, etc.).

III. Biogas Enrichment Techniques

As discussed earlier, biogas enrichment is either possible by removing CO_2 or to some extent H_2S/Siloxane and water removal. Methane enrichment is an emerging concept for high volumetric CH_4 production combined with a conventional biogas plant. The enrichment process is carried out either with in-situ injection of H_2 inside the anaerobic digester or with H_2 injection in a separate reactor called ex-situ, where biogas is upgraded. Figure 3 shows various approaches for improved quality of biogas aimed at increasing the yield of CH_4 have been studied like membrane technology, absorption, scrubbing, biological techniques, hydrogenotropic methanogenesis etc.

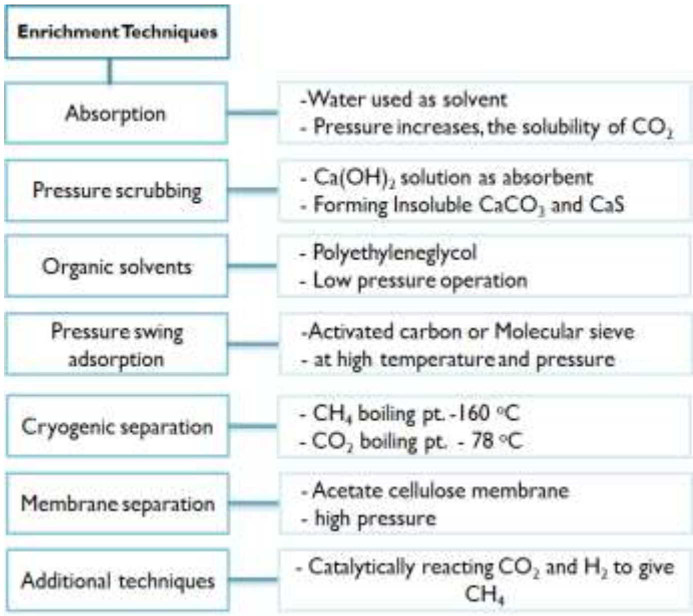

Fig. 3: Various approaches forbiogasenrichment

3.1 Absorption

Various alkyl-amines, commonly known as amines, are used to remove hydrogen sulfide (H_2S) and carbon dioxide (CO_2). Chemical absorption occurs through a reaction between CO_2 and amine-based solvents. Amine scrubbing is a chemical absorption process where solvents like methyldiethanolamine, diethanolamine, or monoethanolamine form a reversible chemical bond with the solute, usually at higher temperatures. The benefits of amine absorption include the complete removal of H_2S, high efficiency, rapid reaction rates compared to water scrubbing, and the ability to operate at low pressure.

3.2 Scrubbing

In water scrubbing, carbon dioxide is primarily absorbed into water, which acts as the solvent to selectively remove CO_2 due to its higher solubility in water compared to methane. This method also effectively removes hydrogen sulfide. Recent research on pressurized water scrubbing emphasizes reducing water usage and applying high pressure, demonstrating a 99.1% reduction in siloxanes and a 9.9% reduction in halogenated compounds.

3.3 Pressure Swing Adsorption (PSA)

Pressure Swing Adsorption (PSA) involves capturing carbon dioxide on a material like activated carbon and regenerating it by lowering pressure. The separation relies on the varying affinities of gases for the adsorbent. This dry separation technology uses porous solids with high surface areas, such as activated carbon, which selectively adsorbs gases through Van der Waals forces. PSA can purify biogas to approximately 97% methane content. Although the adsorbent also captures H2S, this process is irreversible. To prevent damage to the adsorbent, H2S is removed in a separate preliminary step.

3.4 Membranes

Recent advancements have significantly increased the market share of membrane separation over the past three years. Experiments focused on the kinetics of H_2 uptake using pulse H_2 injection for in-situ biogas upgrading, which enhanced the methanogenic mixed culture's adaptation to an H_2-rich environment, ultimately achieving 100% CH_4 enrichment. However, membrane separation has some drawbacks, including relatively low CH_4 and H_2 yields from CH_4 and the high cost of membranes. While some membranes are less expensive, they often suffer from lower yield and purity, as well as membrane fouling, necessitating frequent replacements.

3.5 Cryogenic separation

Cryogenic separation works by utilizing the different condensation and sublimation temperatures of methane and carbon dioxide to separate these gas components into distinct phases. Although the cryogenic technology is technically complex, it results in very high methane purity levels (>99%) with minimal methane losses (<1%). Additionally, it has a low electrical energy requirement, around 5–10%, making it an environmentally friendly approach. Cryogenic valorization relies on the different boiling points of gases to separate CO_2 and CH_4. The biogas is compressed and cooled until the CO_2 condenses into a liquid, allowing it to be separated, while the gas phase becomes concentrated in CH_4.

IV. A Novel Technique: Bio-electrochemical system (BES)

Recently, there has been renewed research interest in electro bioremediation, driven by the rapid development of microbial electrochemical technologies (MET) or bio-electrochemical systems (BES). These systems are unique in that microorganisms directly use insoluble solid electrodes as electron acceptors or donors, enabling the biological oxidation or reduction of organic compounds. This allows for the direct monitoring of organic pollutant degradation through electrical signals (such as current, voltage, or electrode potentials) and enables control by adjusting these variables. Like conventional electrochemical bio-stimulation techniques, BESs do not require the addition of external reagents and offer the potential to directly recover energy as electricity for on-site applications.

A particularly promising aspect of BES technology is the production of methane through the microbial conversion of carbon dioxide with the addition of electrical energy. The first bio-electrochemical production of methane was achieved in 1999 by Park et al., with focused research on methane as the primary product beginning in 2008. In this process, CO_2 can be metabolically reduced to CH_4 by electroactive methanogens using electrons or reducing equivalents, particularly H_2 derived from the cathode. Electromethanogens can either directly accept electrons from electrodes or use bio-electrochemically produced H_2 for CH_4 production as shown in the corresponding reaction eqs 1,2 and 3.

$$CO_2 + 8H^+ + 8e^- \rightarrow CH_4 + 2H_2O \quad (1)$$
$$CO_2 + 4H_2 \rightarrow CH_4 + 2H_2O \quad (2)$$
$$2H_2O \rightarrow O_2 + 4H^+ + 4e^- \quad (3)$$

The concept is based on applying a current between two electrodes (an anode and a cathode) within the anaerobic digestion liquid, typically within a microbial electrolysis cell (MEC). At the anode, organic matter is decomposed, and electrons are transferred to methanogens (such as Methanosaeta and Methanosarcina) by various exoelectrogenic microbial species (primarily Shewanella, Geobacter, and Pseudomonas), facilitating the conversion of biological CO2 into methane (Eq. 4, 5). Methane production in a bio-cathode occurs mainly through two mechanisms: hydrogenotrophic methanogenesis (Eq. 4), where hydrogen serves as an electrochemical mediator—produced either bio-electrochemically or electrochemically—or directly using electrons as a reducing power source (Eq. 5).

$$CO_2 + 4H_2 \rightarrow CH_4 + 2H_2O \quad (4)$$
$$CO_2 + 8H^+ + 8e^- \rightarrow CH_4 + 2H_2O \quad (5)$$

A system can be classified as a BES (bio-electrochemical system) if it includes three essential components: (1) a pair of inert solid electrodes (anode and cathode), upon which the metabolism of a microbial culture (usually a consortium of microbes) depends; (2) an external electrical circuit that connects the two electrodes, typically through a resistive load, power supply, potentiostat, or galvanostat; and (3) an ion mobilization channel or pathway between the electrodes to maintain electro-neutrality or charge balance within the BES.

Optionally, separator materials like ion exchange membranes or non-electrically conductive barrier layers such as glass fiber mesh can be used to keep the two electrodes close together but separated. Redox-active shuttles or mediators can also aid in facilitating exocellular microbial electron transfers within the system. These mediators can be naturally produced by microorganisms (e.g., cobalamins, phenazines) present in the contaminated environment (e.g., humic substances [HS]), or they can be synthetic (e.g., neutral red) and added artificially. Research has shown that placing a pair of electrodes directly inside a digester can improve chemical oxygen demand (COD) removal, biogas production, and methane content. However, this improvement isn't solely due to electro-methanogenesis; it's also influenced by enhanced organic matter hydrolysis, additional electrons supplied by the cathode, and improved syntrophic interactions within the immobilized biofilm. There are two main strategies for integrating electro-methanogenesis with anaerobic digestion (AD). One involves placing electrodes inside a traditional digester, which requires precise control, while the other uses a BES as a post-treatment to AD, simplifying the operation of both systems and offering a more versatile setup to optimize both processes.

BESs have been explored for their potential in various applications, especially in wastewater treatment, where they can simultaneously produce bioenergy. However, the performance of these complex systems, in terms of COD removal and energy or chemical production, is influenced by numerous factors, including the type of wastewater (substrate), electrode material, separator material, inoculum type, reactor configuration, and mode of operation.

To enhance the system's performance, various bio-electrochemical treatment and enhancement measures can be applied. These include adjusting pH, temperature, or moisture levels; acclimating anode microorganisms to magnetic fields; performing anaerobic treatment in the anode and aeration in the cathode; adding external nutrient sources (e.g., glucose, vitamins, inorganic salt ions) or co-metabolic substrates (e.g., acetate, pyruvate, glucose, sucrose); altering the conductivity of the environmental media or electron transfer activity (e.g., adding activated carbon, carbon, gravel, salt, or external electron mediators); increasing current by changing the external resistance; or setting the potential via a potentiostat. These measures can modify the properties of the environmental media, thereby influencing microbial activity, species composition, and structure, which in turn affect contaminant removal and electricity production in the BES.

V. Role of Electrode

In recent years, various electrode materials have been explored and developed to enhance the performance of microbial fuel cells (MFCs) and microbial electrolysis cells (MECs) while also reducing reactor costs. In these bio-electrochemical systems (BESs), electrodes serve not only as conductors but also as habitats for electrogenic microbes. Therefore, ideal electrode materials should have specific surface characteristics, such as good biocompatibility, a high specific surface area, significant surface roughness, and the ability to efficiently transfer electrons between microbes and electrodes. Given these requirements, carbon-based materials are often preferred for use as anodes.

The three most commonly used carbonaceous electrode materials are carbon paper, carbon cloth, and graphite brushes. Carbon paper is thin, firm, and slightly brittle, with a relatively smooth surface. In contrast, carbon cloth is more flexible and porous. Graphite brush, a fiber fabric much thicker than the other two materials, has a brush-like configuration that provides more surface area (though less than carbon cloth) for microbial attachment. However, the high cost of these carbon-based materials, particularly carbon cloth, poses a significant barrier to the practical application of BESs. Additionally, the high electrical resistivity of these materials can lead to significant ohmic losses in large-scale systems.

VI. Conclusion

The global energy demands continue to rise, driven by population growth and industrialization, the need for sustainable energy solutions becomes increasingly urgent. While traditional methods for biogas upgrading have been effective, they are often costly and energy-intensive. Emerging technologies like bio-electrochemical systems (BES) present a promising alternative, offering the potential for more efficient and sustainable biogas enrichment. By enhancing methane production and integrating with anaerobic digestion, BES technology could play a crucial role in addressing energy scarcity and reducing environmental impacts. Continued research and innovation in BES technologies are essential to overcoming current challenges and achieving practical, cost-effective solutions for renewable energy production.

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