

# Technologies for Biogas Upgrading to Biomethane: A Review

J. M. Mutunga\*, H. M. Ndiritu, M. Hawi, and P. Oketch

**Abstract**— Biogas production from anaerobic digestion of organic waste has gained significant attention globally in recent years as it addresses both energy and environmental challenges. It is primarily used for cooking, lighting, and heating purposes. Biogas upgrading technologies have been developed to increase the scope of its application to natural gas grid injection and as a substitute fuel in the automotive industry by removing biomethane contaminants in biogas which include; carbon dioxide ( $\text{CO}_2$ ), hydrogen sulphide ( $\text{H}_2\text{S}$ ), water vapour ( $\text{H}_2\text{O}$ ), nitrogen ( $\text{N}_2$ ) and oxygen ( $\text{O}_2$ ). Both physico-chemical (sorption and separation), and biological processes exist for specific applications. High energy consumption during biogas upgrading process is a concern to environmental and economic sustainability. This work evaluates existing and emerging biogas upgrading technologies with a special emphasis on adsorption technique for  $\text{CO}_2$  and  $\text{H}_2\text{S}$  removal. Even with the high results of up to 99% biomethane ( $\text{CH}_4$ ) purity, after  $\text{CO}_2$  and  $\text{H}_2\text{S}$  removal using biogas upgrading technologies, their optimization is inevitable in strengthening biogas as a reliable renewable energy alternative in the energy sector. Temperature based regeneration of adsorption technique, using activated carbon as adsorbent, and relying on renewable energy supply is recommended. Its compact nature, ease of operation, and promising ability to simultaneously remove carbon dioxide and hydrogen sulphide makes it a sustainable solution to some of the challenges faced in the biogas upgrading field.

**Keywords**— Adsorption, Biogas contaminants, Biogas upgrading, Biomethane.

## I. INTRODUCTION

THE heavy reliance on fossil fuels globally as a source of energy is linked to the rising levels of greenhouse gases in the atmosphere, and consequently, climate change [1]. Given the increasing trend in global energy demand projections, renewable energy sources are a reliable source due to their continual ability to self-replenish. As a result, renewable energy generation has received a lot of attention, especially among researchers, as an alternative source of energy.

Wind, solar, and biomass energies have greatly dominated the renewable energy sector. The increasing exploitation of wind and solar energies provides a cheaper alternative energy

source but these systems are characterized by highly fluctuating and poorly predictable production profiles [2]. Biomass energy on the other hand is capable of producing a constant base load and even balancing out the supply-demand variations in the sector. Its utilization is also significantly independent of geographical location and seasons [3].

Energy can be harnessed from biomass either by gasification to produce syngas and hydrogen, or by anaerobic digestion to produce biogas. Differences related to the heating values of biogas (20-26 MJ/m<sup>3</sup>) and syngas (10-18 MJ/m<sup>3</sup>) suggest that the use of biogas is preferred over syngas to achieve high energy production [4]. Biogas is a product of anaerobic digestion (AD) that involves bacteria breaking down biological materials in the absence of oxygen. The main stages of anaerobic digestion process include hydrolysis, acidogenesis, acetogenesis, and methanogenesis, as shown in Fig.1 [5]. Biomethane ( $\text{CH}_4$ ) and carbon dioxide ( $\text{CO}_2$ ) are the main constituents of raw biogas with compositions of (50 to 65 % (v/v)) and (35 to 55 % (v/v)) respectively, saturated with water vapor at given temperatures. Other important minor constituents include oxygen ( $\text{O}_2$ ) from air, nitrogen ( $\text{N}_2$ ), hydrogen sulphide ( $\text{H}_2\text{S}$ ), carbon monoxide (CO), and ammonia ( $\text{NH}_3$ ) [6].

Energy content in biogas can be harnessed primarily by combustion in a gas engine or a suitable burner to produce electric and thermal energy respectively. Electrical power from biogas is not economically competitive compared to other sources, with the electrical efficiency of modern gas engines ranging between 33% and 40%. When used as a source of thermal energy, the efficiency ranges between 83% and 88% [3]. Upgrading of raw biogas to stipulated specification has seen an increase in its application as a renewable natural gas substitute, and as a local fuel when compressed.

Technologies focused on  $\text{CO}_2$  removal, the major non-combustible component of raw biogas, have greatly evolved over the years. Its presence in raw biogas limits application to low-quality energy applications such as lighting and cooking. During the biogas upgrading process,  $\text{CO}_2$  can either be

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removed and used for various applications such as mineralization and synthesis of polycarbonates, or converted to  $\text{CH}_4$  by reaction with  $\text{H}_2\text{S}$  [7]. Most of the mature technologies in biogas upgrading process have applied the former.  $\text{H}_2\text{S}$  removal is also critical since in the presence of water vapor it becomes corrosive reducing the lifespan of metallic equipment such as valves, engines, and pipes. Elimination of the other minor contaminants from raw biogas depends on its utilization

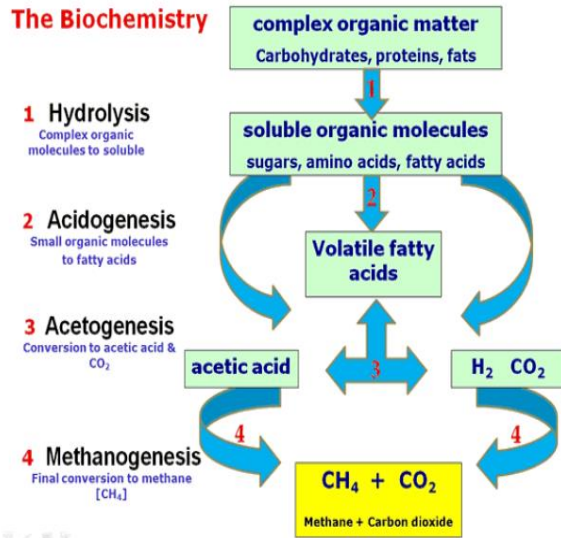


Fig.1 The anaerobic digestion biochemical conversion pathways [5]

With the increasing advancement in biogas upgrading process, evaluation of the utilized technologies is essential. Most of the physical and chemical biogas upgrading processes are energy intensive. This has seen adoption of biotechnologies in the upgrading process, and or integration with other renewable energy sources such as solar and wind as energy sources for the upgrading process [8]. This paper critically reviews and discusses the state-of-the-art technologies for removal of  $\text{CO}_2$  and  $\text{H}_2\text{S}$  from raw biogas, with a special focus on the adsorption technique. Significant technological breakthroughs in the field within the past decade are reviewed.

## II. BIOGAS UPGRADING TECHNOLOGIES

Several technologies for biogas upgrading have been studied and implemented at industrial scale with the aim of encouraging biogas energy uptake. These processes can be categorized based on the point of contaminant removal during the biogas production process as; pre digestion, during digestion and post digestion upgrading. Classification based on the contaminant being removed has also been adopted as it clearly represents process evolution with minimal external influence. This is adopted for this work, with a specific focus on  $\text{CO}_2$  and  $\text{H}_2\text{S}$  contaminants. Recent studies are also adopting classification based on simultaneously removal of more than one biogas contaminant.

### A. Carbon Dioxide ( $\text{CO}_2$ ) Removal

Majority of the mature, industrial scale, and commercially available biogas upgrading technologies have focused on  $\text{CO}_2$  removal from raw biogas. Being the highest non-combustible gas by composition, and significantly affecting the average calorific value of biogas (raw biogas,  $24 \text{ MJ/m}^3$ ; biomethane,  $36 \text{ MJ/m}^3$ ) have greatly contributed to the much attention it has received [9]. These techniques rely on physical, chemical, and thermodynamic variations of gas properties in separation of the raw biogas components. Sorption (absorption and adsorption) and separation (membrane and cryogenic) techniques are some of the available technologies for commercial use. Recently,  $\text{CO}_2$  removal using biological approaches has been proposed. However, most of them are in the research and development, and pilot stages. Governing principles and current status of these technologies are discussed below with a more in-depth analysis of the adsorption process.

Absorption technique is the most common, well developed, and widely implemented approach in biogas upgrading, accounting for about 63% of the global market [10]. Counterflow of the absorbent liquid and raw biogas in a column filled with packings increases their contact area allowing for maximum  $\text{CO}_2$  to dissolve in the absorbent, being more soluble than  $\text{CH}_4$  [11]. Depending on the governing principles, absorption can be classified into physical and chemical processes. Physical absorption is dependent on the physical properties of both the gas and the liquid such as temperature, pressure, and solubility. Water and polyethylene glycol are some of the commonly used absorbents for this process. Chemical absorption is a chemical process that is irreversible and commonly uses amines and alkali solutions as the absorbent. Even with the high  $\text{CH}_4$  purity of up to 97%, the high installation and operation cost involved such as pumps and compressors running limits its application in small scale setups. The high water flow rates of up to  $100 \text{ Nm}^3/\text{h}$  required in upgrading biogas of up to  $200 \text{ m}^3/\text{h}$  depending on the working pressure further limits its application in arid and semi-arid areas where water is scarce [12].

Membrane technology for biogas upgrading has also strongly been adopted given that it is the core technology used in natural gas industry to remove contaminants, with a market share of up to 10% [12], [13]. Separation is based on the selective permeability properties of membrane materials such as polymeric (polyimide, polysulfone, polyetherimide), inorganic/non-polymeric (zeolites, silica, carbon molecular sieves), and composites of the same [14]. An ideal membrane in raw biogas purification should have a high permeability difference between  $\text{CO}_2$  and  $\text{CH}_4$ , be able to withstand the high operating pressures of 5 to 20 bar, and be corrosion resistant due to the presence of  $\text{H}_2\text{S}$  and  $\text{NH}_3$  in raw biogas. During the separation process,  $\text{CO}_2$  and other gas contaminants permeate through the membrane, with methane being retained. Prior to membrane separation, raw biogas is passed through a filter to remove water vapour that would otherwise negatively affect the performance of the membrane [15]. The performance of this

technology greatly depends on the system configurations put in place such as the number of pressure stages and loops adopted. To minimize on methane loss during the upgrading process, compression of raw biogas is inevitable [16]. Adoption of the multistage approach has increased  $\text{CH}_4$  purity to 96%, but greatly affect the biomethane output pressure which could limit its application [17].

Contents of raw biogas liquify at different temperature and pressure conditions making it possible to separate them by compression and cooling. Upgrading of raw biogas using the cryogenic technique involves first drying the gas to prevent freezing during the cooling process. It is then compressed at multiple stages up to 8,000 kPa and then cooled to  $-45^\circ\text{C}$  where the condensed  $\text{CO}_2$  is removed. The biogas is further cooled to  $-55^\circ\text{C}$ ; afterward expanded to 800 - 1,000 kPa reaching a temperature of  $-110^\circ\text{C}$ . In these conditions, there is a gas-solid phase balance, with the solid phase being  $\text{CO}_2$  and the gaseous phase containing more than 97%  $\text{CH}_4$ , that is collected and heated before leaving the process [15], [18]. The cryogenic purification technique has been able to achieve a very high  $\text{CH}_4$  purity of 97 - 98% with less than 2% loss. It is also environmentally friendly as it does not utilize chemicals that would negatively affect the ecosystem upon disposal. It however requires huge investment and operation costs, with high energy demand to power the cooling systems. Furthermore, the technology is still under development with a global market share of 0.4% [10], [19].

Biological reduction of  $\text{CO}_2$  from raw biogas can be categorized based on the point of occurrence as either during or post anaerobic digestion. Hydrogen ( $\text{H}_2$ ) assisted  $\text{CO}_2$  bioconversion has been implemented in both cases. Hydrogenotrophic methanogens use  $\text{CO}_2$  as their carbon source and electron acceptor, with  $\text{H}_2\text{S}$  being the electron donor in the energy-yielding reaction that gives  $\text{CH}_4$  and water vapor as the products [20], [21].  $\text{H}_2$  obtained mainly by water electrolysis is injected into the digester and stirred to facilitate the reaction process. Implementation of the technology during the anaerobic process has the advantage of reduced equipment cost. Its drawbacks include the high flammability of hydrogen, anaerobic conditions are required for biogas generation, and the negative impact excessive mixing could have on biogas production and design of the digester. This makes  $\text{CO}_2$  bioconversion post anaerobic digestion a good alternative [8]. The technology is currently under research with most of the prototypes being lab scale.

### B. Hydrogen Sulphide ( $\text{H}_2\text{S}$ ) Removal

$\text{H}_2\text{S}$  is formed during the microbiological reduction of sulfur-containing compounds (sulfates and amino acids) during the anaerobic digestion process. Its presence not only affects the quality and quantity of biogas generated, but also produces harmful environmental emissions and corrodes metallic parts of the biogas upgrading systems [22]. For these reasons,  $\text{H}_2\text{S}$  is mostly the first raw biogas contaminant to be removed. Its concentration varies depending on the biomass feedstock used and the biogas production process; ranging between 50 to 3000

ppm, or higher [23]. The recommended concentration of  $\text{H}_2\text{S}$  in biogas that finds a similar application as natural gas is 16 ppm or less [24]. This necessitates the removal of  $\text{H}_2\text{S}$  during the biogas upgrading process.

Most of the fully developed physicochemical technologies used in  $\text{CO}_2$  removal from raw biogas such as absorption, adsorption and membrane technologies can be used in  $\text{H}_2\text{S}$  removal. They apply the same working principles discussed in the previous section. Precipitation of  $\text{H}_2\text{S}$  in the digester using  $\text{Fe}^{2+}$  and  $\text{Fe}^{3+}$  has been widely studied. It has proved efficient for applications having high  $\text{H}_2\text{S}$  concentrations ( $> 200$  ppm). Its major drawback is the high chemical cost incurred during the process [15]. Physical absorption using either water or organic solvents is a common approach of removing low  $\text{H}_2\text{S}$  concentrations from raw biogas. This is attributed to the simplicity of its configuration. The approach is more competitive when  $\text{CO}_2$  is also simultaneously removed during the process [18].

Membrane thickness and tortuosity, operating pressure and temperature, and flow design are key variables that affect efficiency of membrane technology in  $\text{H}_2\text{S}$  removal. Use of hollow fiber membrane made of polymeric material was studied [25]. Increased pressure ratio across the membrane of up to 8 bars increased  $\text{H}_2\text{S}$  selectivity in the retentate and  $\text{CH}_4$  purity, but resulted in decreased  $\text{CH}_4$  recovery. While simultaneous removal of  $\text{CO}_2$  and  $\text{H}_2\text{S}$  was achieved, optimization of working pressures that allow for maximum separation of the biogas contaminants is essential. In addition, more research on the life time of the membrane is curtail to determine techno-economic feasibility of this technology in the industry.

The use of biotechnology in removal of  $\text{H}_2\text{S}$  from raw biogas is currently being implemented at full scale having equal performance and less operation cost when compared to physicochemical approaches [8]. Its application is mainly in in-situ configurations. Naturally occurring sulfur oxidizing bacteria such as lithoautotrophic bacteria is used in the desulfurization process. Both  $\text{O}_2$  and  $\text{NO}_3^-$  are effective electron acceptors in the treatment of  $\text{H}_2\text{S}$  but can achieve 100% removal efficiency only when the concentrations in raw biogas are below 2000 ppm [26], [27]. Controlled air supply is a less costly approach of introducing  $\text{O}_2$  electron to the digester. This approach can however have a negatively impact on the calorific value of the gas due to the increased volume of nitrogen gas contained in air.

### III. ADSORPTION TECHNOLOGY

Adsorption technology is based on the different molecular characteristics of the different gas contaminants in raw biogas and their varied affinity towards adsorbent materials. It has proved to be effective for both  $\text{CO}_2$  and  $\text{H}_2\text{S}$  separation in raw biogas with studies analyzing efficiency for simultaneous removal of the gases [11]. Even with numerical and experimental research having been conducted in the area, and as a result it is being considered a mature technology in the biogas upgrading field, ongoing research is mainly focused on

optimizing the process. IPSEpro, gPROMs and Aspen (Hysys, Plus, and Adsorption) are some of the software commonly used in simulation of the adsorption process [28]–[30].

The main steps in the adsorption process are adsorption, adsorbent regeneration, and desorption/purging. Fig.2 shows the major classification of adsorption techniques based on the different regeneration approaches adopted. Pressure and temperature are the main gas properties that affect the system regeneration performance. Use of multiple adsorption columns (up to nine) in parallel configuration has ensured continuous operation of the raw biogas upgrading process [31]. In addition, the gas cyclic approach adopted in most adsorption processes has increased efficiency, reduced energy consumption, and reduced methane losses.

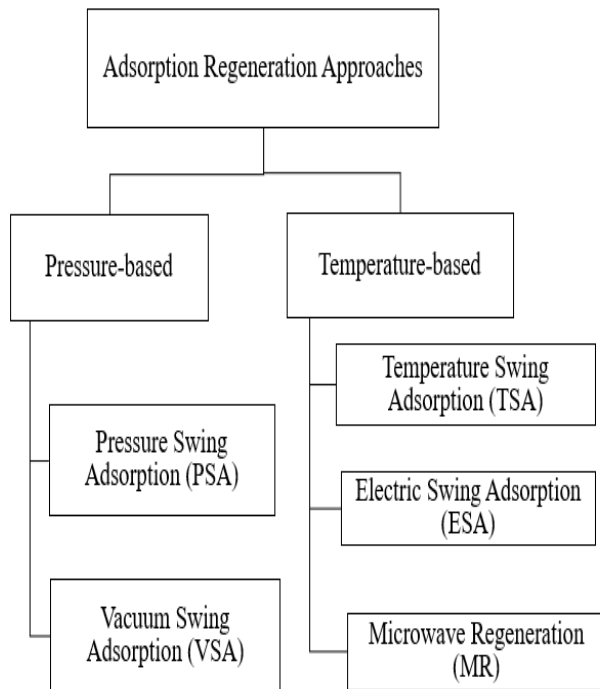


Fig. 2 Classification of adsorption regeneration approaches

Zeolites (classified according to their pore size as zeolite 13x, zeolite 5A, zeolite 4A), carbon molecular sieve, silica gel, and activated carbon are common adsorbents used in biogas upgrading by adsorption. This is due to having higher  $\text{CO}_2$  solid-gas partition efficiency when compared to that of  $\text{CH}_4$  [32]. These adsorbents are commercially available mainly in granular and powder form. However, recent studies using 3D printed adsorbent layers have demonstrated lower  $\text{CO}_2$  loading capacities necessitating optimization of their mechanical and composition properties [33]. Zeolites have high  $\text{CO}_2$  loading capacities at low pressure when compared to activated carbon, that in addition has better electrical conductivity properties, making them favorable for temperature-based regeneration [34]. Use of silica gel granules in pressure-based adsorption regeneration process yielded satisfactory results of 98%  $\text{CH}_4$  purity when compared to other adsorbents such as zeolites and activated carbon [35].

### A. Pressure-based Regeneration

Pressure-based regeneration of adsorbents for biogas upgrading is based on the theory that pressurized gases are attracted on solid surfaces and reducing the pressure releases the gases. Pressure Swing Adsorption (PSA) columns are often operated at high pressures of 4 to 10 bar to selectively retain  $\text{CO}_2$ , with working pressures above 10 bar drastically reducing the  $\text{CO}_2/\text{CH}_4$  separation efficiency [36]. Most recent multiple steps adopted for this upgrading process included; adsorption, equalization depressurization, blowdown, purge, equalization repressurization, and pressurization [31]. Pressure-based adsorption process has the advantage of equipment compactness; therefore, small, compact and modular units can be easily fabricated for small-scale applications. The high energy consumption is the main challenge facing this approach of biogas upgrading technique.

Vacuum Swing Adsorption (VSA) applies similar working principles to that of PSA with the major difference being that desorption process occurs under vacuum conditions. The high capital cost of equipment required to achieve deep vacuum levels has contributed to the adoption of minimum desorption pressure of 0.1 bar [31], [37]. Recent studies have combined both PSA and VSA principle, applying Vacuum Pressure Swing Adsorption (VPSA) principles in biogas upgrading process as shown in Fig.3.  $\text{CH}_4$  purity of 98% reported was greatly influenced by the short adsorption time, low desorption pressure, and high purge to flow ratio. In addition, use of carbon molecular sieve adsorbents in the VPSA process has better  $\text{CH}_4$  recovery rates when compared to zeolite 13x [31].

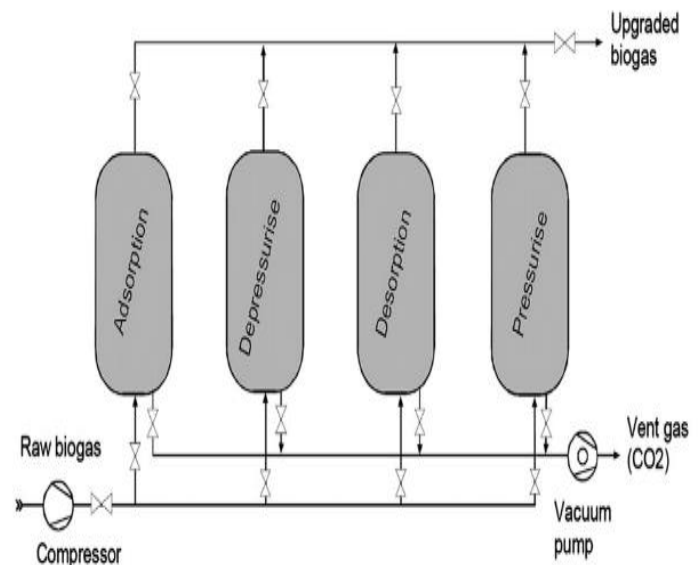


Fig. 3 Set-up of (vacuum) pressure swing adsorption [15]

### B. Temperature-based Regeneration

The main steps implemented in this approach include; adsorption, heating, purging and cooling. During the adsorption process, raw biogas is fed into the adsorption column to allow for separation, and the process stopped before saturation is reached. Upon saturation of  $\text{CO}_2$  on the adsorbent material,

mostly represented by the creation of a film on the surface of the adsorbent, adsorption process is stopped to allow for regeneration. In the case of Temperature Swing Adsorption (TSA), a fluid is heated then passed through the column to facilitate regeneration, while for Electric Swing Adsorption (ESA), the column is directly heated by passing electric current through it as illustrated in Fig.4 and Fig.5 respectively. The use of inert gases such as helium and nitrogen as purge gases is common among most temperature-based regeneration processes. Reuse of a fraction of the upgrade biogas (biomethane) as a purge gas is an approach recently adopted to reduce the overall operation cost. Chemisorbents are more preferred when compared to physisorbents in temperature-based adsorption processes due to their rapid cyclic and high working capacities at elevated temperatures.

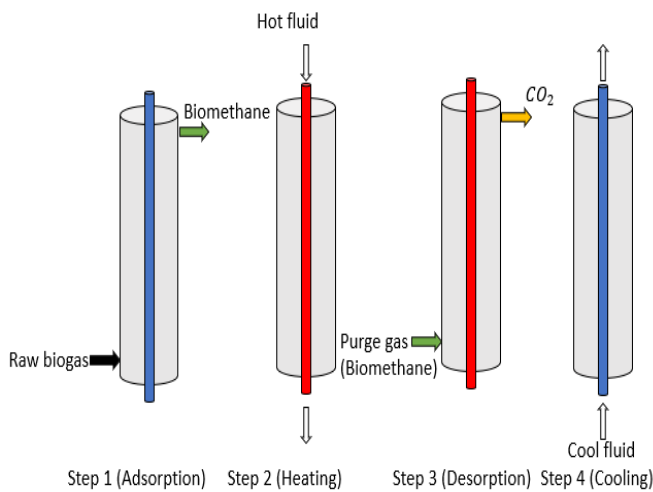


Fig. 4 Schematic diagram of TSA cycle with four steps

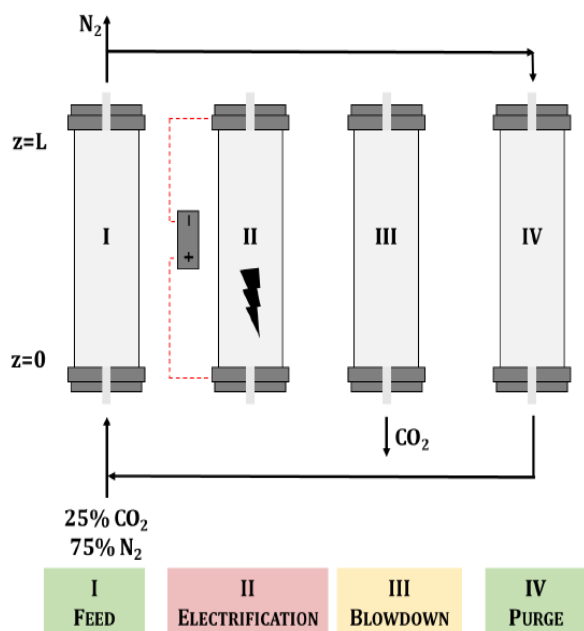


Fig. 5 Schematic diagram of ESA cycle with four steps [38]

The application of TSA technology is more feasible when

energy is harnessed from post combustion gases (waste gases). As a result, its application is common for CO<sub>2</sub> capture from flue gases with limited application in biogas upgrading process [39]. The use of a heat pump for the regeneration process has been tested with high energy requirements of 2.8 MJ/kg of biomethane [40]. The long period of time required during the heating and cooling steps of the process contributes to thermal aging of adsorbents which greatly reduces the process efficiency [41]. For feasible application of TSA in biogas upgrading, a sustainable energy source is required.

ESA is considered a second-generation technology for CO<sub>2</sub> separation. High electric powers (voltage and current) allow for faster heating of columns, reducing the time required during the heating step. While copper, aluminum, and brass are some of the electrodes proposed for electric conductivity (indirect ESA), use of adsorbents that are good conductors of electricity (direct ESA) have proved to increase the overall system efficiency as it reduces heat loss due to contact resistance between the electrode and adsorbent [42], [43]. Similar to TSA, ESA has mainly been studied for CO<sub>2</sub> capture from flue gases post combustion, with limited application in CO<sub>2</sub> separation from raw biogas [42]. While some research work have tried to mimic the composition profiles of raw biogas by synthetic mixing of CO<sub>2</sub> and CH<sub>4</sub>, use of ESA in upgrading raw biogas obtained from anaerobic digestion is necessary to clearly understand its viability in biogas upgrading [44].

Microwave regeneration technology (MR) utilizes electromagnetic energy that is converted into thermal energy in the adsorbent bed. Microwave energy is delivered directly to the adsorbent by varying magnetic field, as opposed to conventional heat transfer processes such as conduction and convection that require a medium. As a result of the direct heating, low purge gas flow rates are required during the purging step making it economically viable. The major advantages of MR include low energy consumption, dependency only on the adsorbent dielectric properties, and high adsorbate removal capacity. While the technology has been tested for N<sub>2</sub> and CO<sub>2</sub> separation with energy efficiency of up to 75%, there is limited application of the same in biogas upgrading [45]. It is a feasible alternative for regeneration during biogas upgrading process given its environmental sustainability nature when the electric energy is harnessed from renewable energy sources such as wind and solar.

#### IV. CONCLUSION

Physicochemical technologies for raw biogas upgrading to biomethane have demonstrated great performance with high levels of CH<sub>4</sub> purity achieved. Biotechnology is also a promising alternative being a more environment-friendly option. Equipment compactness, safety, and simplicity of operation are the major advantages associated with adsorption process. However, the high energy requirements and costly chemicals required for these processes are challenges hindering its application in raw biogas upgrading processes.

Temperature-based regeneration for adsorption processes is

a promising approach that could see full exploitation of biogas as a renewable energy source if the energy is harnessed from other renewable energy sources such as solar and wind. Activated carbon is a preferred adsorbent given its better thermal properties when compared to zeolites. Enhancement of its properties could allow for simultaneous removal of carbon dioxide and hydrogen sulphide which would make the technique more economical. In addition, optimization of biogas upgrading processes will not only ensure energy security, but also encourage waste management, and climate change mitigation globally.

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