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Review

Biogas Valorisation to Biomethane for Commercialisation in South Africa: A Review

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Abstract: Biogas consists of mainly methane, as a source of energy, and impurities such as carbon dioxide, hydrogen sulphide, water, and siloxanes. These impurities, such as hydrogen sulphide, reduce the biogas energy content and corrode equipment that store, transport, or utilise biogas. Several reviews on upgrading biogas to biomethane have been published, but minimal focus has been put on upgrading biogas for commercialisation in South Africa. Thus, this study reviewed biogas upgrading techniques in South Africa to put together information on activities and experiences on biogas valorisation to enhance the chances for different stakeholders to learn and build on from local experiences. To capture all relevant information, literature from the past 10 years was retrieved from online databases and government, municipality, and companies' websites and institutional repositories. The review covered the sorption, separation, and in situ techniques that are globally used for upgrading biogas. The status of the biogas sector and the upgrading activities that occur in the country with their cost, energy, and environmental impacts were given in detail. It is estimated that a total of 3 million Nm^3d^{-1} of biogas can be produced in the country from biogas substrates. Thus, researchers and entrepreneurs are encouraged to collaborate to utilise the abundant resources used for biogas production to enhance the commercialisation of biomethane.

Keywords: biogas; biogas valorisation; biogas upgrading; biomethane; sorption; separation; methane purity; organic waste

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1. Introduction

The constitution of the Republic of South Africa (RSA) stipulates that everyone is entitled to a safe environment for their health and well-being [1]. The negative environmental impacts of poor waste management have led the South Africa National Waste Management Strategy (NWMS) 2020 to focus on the reduction of waste going to landfills by 70% through reuse, recycling, recovery, and alternative waste treatment in the next 15 years [2]. Anaerobic digestion (AD), whose benefits are shown in Figure 1, has established itself as an alternative technology to rid of organic waste that is diverted from landfills, wastewater, and farming biomass residue while producing biogas, an alternative source energy, whose global consumption has increased by 90% from 65 GW in 2010 to 120 GW in 2019 [3]. Biogas has the potential to enhance sustainable development, reduce energy related pollution, enable communities to manage their waste in a beneficial manner, contribute to the abatement of climate change, and stop firewood-linked deforestation. However, biogas applications are limited by the presence of impurities, namely 25–45% carbon dioxide (CO_2), 20–20,000 ppm hydrogen sulphide (H_2S), 2–7% water (H_2O), traces of ammonia (NH_3), siloxanes ($((CH_3)_2 2SiO)_n$), oxygen (O_2), carbon monoxide (CO), nitrogen (N_2), and volatile organic compounds (VOCs) [4,5].

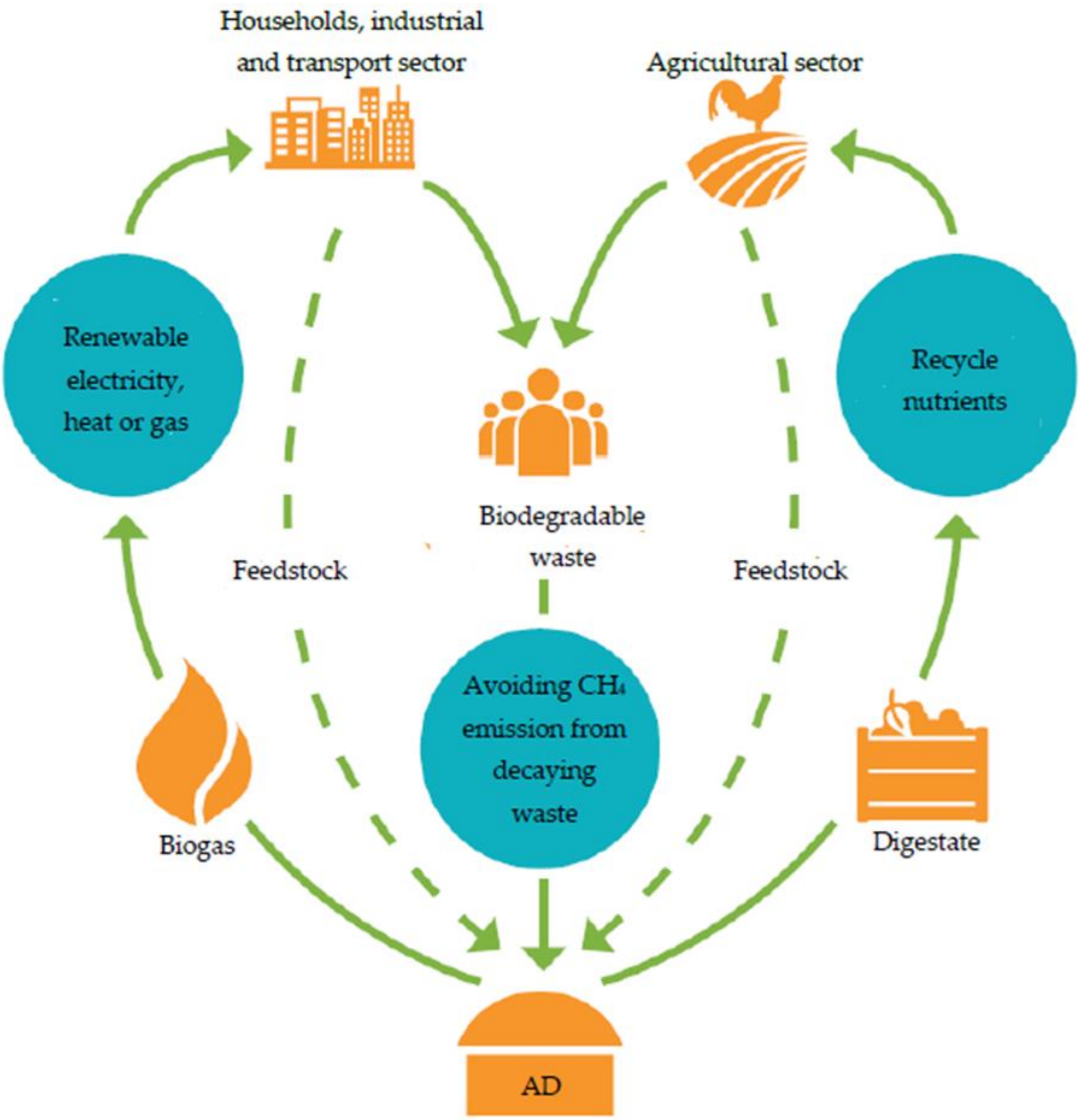

Figure 1. Benefits of anaerobic digestion. Adapted from [6].

The presence of CO_2 decreases the laminar flame speed, combustion efficiency, flammability limits, calorific value, rate of burning, and range of flame stability [7]. It is not easy to ignite biogas at a Reynolds number above 1200 because of the decreased flammability limit. The higher the CO_2 concentration in biogas, the lower the burning velocity and the lower the energy content. Thus, raw biogas is only suitable for low energy demanding applications, such as cooking and lighting. It lowers the biogas density, which results in the increased frequency of refilling fuel tanks for vehicles utilising biogas [8]. Additionally, it forms dry ice on valves, which makes it difficult to store biogas in containers, resulting in limited uses and transportation.

H_2S forms salt in machines by reacting with metals such as lead. It also reacts with H_2O to form sulphur dioxide, and further reaction results in the formation of sulphuric acid, which causes the wearing/corrosion of appliances that use, store, or transport biogas [7]. H_2O accumulates and condenses on surfaces and parts of transporting pipes and equipment that use biogas, where it causes rusting. H_2O also react with NH_3 to form ammonium hydroxide, which corrodes some metals, especially aluminium and copper. NH_3 also reacts with oxygen to form nitrous oxide, which has a global warming potential that is 273 greater than that of CO_2, making it a great contributor to climate change [9].

Nyamukamba et al. (2020) gave a detailed account of siloxanes, which they described as the most harmful trace compounds found in biogas since they are converted to silica that damages gas engines, turbines, or any other equipment that utilises the biogas [10]. During the combustion of biogas, the siloxanes react with oxygen to form silicon dioxide, which forms a layer on the wall surfaces of engine components, for example, combustion chambers, spark plugs and valves. Silicon dioxide is an abrasive electrical and thermal insulator, which causes the wear and tear of the combustion chamber cylinder surface and the impairment of lubricant distribution. This reduces the engines maintenance interval and limits their life, resulting in increased operational costs. Thus, it is advisable to use clean gas; although, different equipment have different tolerances for siloxanes. If released into the atmosphere, silicon dioxide is dangerous to human beings, as it can cause cancer.

The concentration of CH_4, which determines the calorific value of the biogas, is affected by the type and source of feedstock used and the digester operating conditions [11]. To add value to biogas, the impurities must be removed. The removal of harmful substances prevents the degradation of equipment that uses the biogas. The removal requirements of major harmful substances from biogas for various biogas applications are given in Table 1. If not removed, the unwanted substances have varying effects, such as the fouling and corrosion of appliances and equipment and lowering the energy content of the biogas.

Table 1. The removal requirement of major harmful substances from biogas for various biogas applications.

Biogas Application	H_2S	H_2O	CO_2	Siloxanes
Boiler	Recommended <1000 ppm	Not required	Not required	
CHP Transport fuel	Required 542–1742 ppm Required <5 ppmv	Required Required	Required Required	Required 9–44 ppm <21 ppm
Kitchen stove	Required <10 ppm	Not required	Not required	Not required
Natural gas pipeline	Required <4 ppm	Required	Required	
Liquid biomethane	Required <4 ppmv	Required < 1 ppmv	Required < 25 ppmv	

Adapted from [8,12].

There is growing interest in biogas valorisation to biomethane, as shown by the increasing number of published articles in the Scopus database since 1995 that use upgrading, scrubbing, cleaning, valorisation, or compression within 10 words from biogas and that focus on the environment (Figure 2).

Upgraded biogas (biomethane), which can have an energy value up to 36 MJm^{-3}, can be used to substitute fossil fuels in heat and electricity generation and in the transport sector [13]. This would reduce the emission of greenhouse gases (GHG) into the atmosphere by avoiding the burning of fossil fuels, thereby preventing old carbon stored in them from releasing into the atmosphere. Some developed countries have established sustainable biogas systems that have processes for waste pre-treatment, environment protection, the valorisation of material, heat, and/or electricity production, and biofuel production. Since AD systems are dispatchable, the use of biogas can solve the problem of intermittent energy from other renewable energy sources, such as solar and wind.

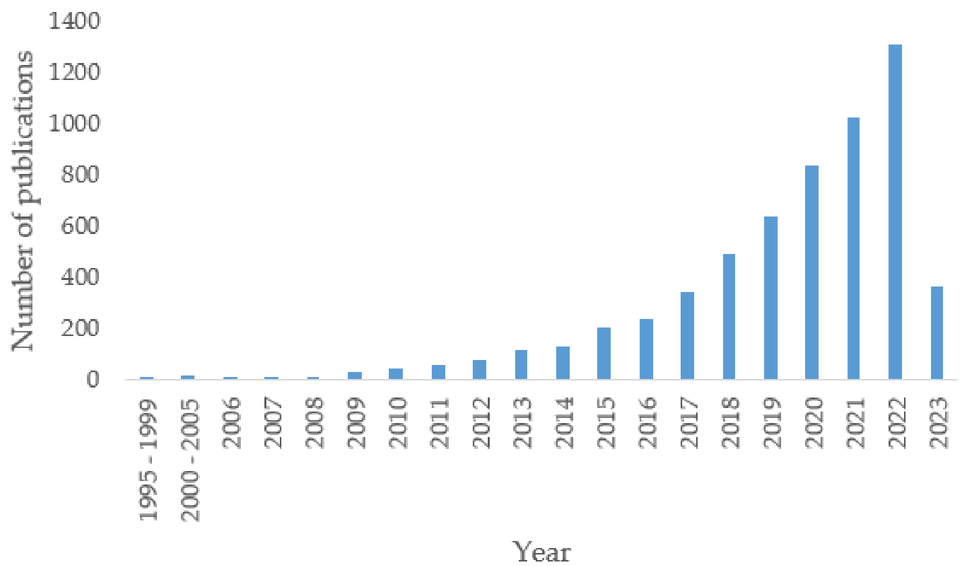

Figure 2. Number of research articles published per year with search query: (upgrading OR scrubbing OR cleaning OR valorisation OR compression W/10 biogas AND (LIMIT-TO (SUBJAR-EA, "ENVI")). Source of data [14].

Despite the progress on the international scale and the presence of more than 700 installed biodigesters in the country [15,16], the focus on the valorisation of biogas to biomethane is still very low in South Africa. Several biogas digesters have stopped operations because farmers do not have the experience or the backing from biogas specialists to improve the energy value of the biogas, and of the 700 biogas plants that were installed by 2017, only 300 were operational [17]. Several biogas plants are registered with the South Africa Biogas Industry Association (SABIA), but they mainly use biogas for electricity production. Only one plant, in Athlone, Cape Town, valorises biogas to biomethane at a commercial scale [18]. Several reviews on biogas upgrading in developed countries have been published, but minimal focus has been put specifically on upgrading biogas to biomethane for commercialisation in South Africa. Consequently, this study reviewed biogas upgrading techniques in South Africa to put together information on activities and experiences on the upgrading of biogas to enhance the possibility of different stakeholders learning from local experiences. Thus, this study aimed to answer the following questions: (1) What methods are being used for the valorisation of biogas in South Africa? (2) What is the cost and energy implication of biogas valorisation methods? (3) What is the national or environmental significance of the valorisation of biogas? Answers to these questions are useful to different stakeholders, such as entrepreneurs in the energy and waste management sectors and policy makers who need to decide on what technologies to use and develop informed policies, respectively. It is hoped that an understanding of biogas upgrading processes may result in the increased uptake of biogas technology in the country.

2. Methods

To obtain an in-depth understanding of the current status of the valorisation of biogas to biomethane for commercialisation in South Africa, this study carried out a detailed integrative literature review, aiming to add value to biogas via biomethane. The analysed literature was retrieved from online databases, namely Scopus, Science Direct, Sematic Scholar, Google Scholar, and SA Media (SABINET). For the Scopus database, the following search terms were used: scrubbing, cleaning, valorisation, and compression within 10 terms from biogas. The same search terms were within 10 terms from South Africa for upgrading techniques that were tried in South Africa. Additionally, government and municipality websites were searched. Although Google and Semantic Scholar did not provide a clear search refining criteria, they were included because they capture a wide range of upcoming journals, which increases the chances that even the tiniest research or activity conducted by various institutions can be captured. SABNET, government, and municipality websites and institutional repositories were included to avoid concentrating on journal articles that only target the scientific community. For all the database and websites, the evaluated publications and documents were all published in the last 10 years to reduce the chances of capturing activities that are no longer active.

An overview of common methods that are currently used for commercial biogas upgrading is presented. The main focus was on mature or near-maturity technologies whose upgrading equipment can be found on the market. Having established a brief state of the upgrading techniques, more focus is given to the local state, where the technologies that are being tried in the country are reviewed. Lastly, the cost, energy, and environmental implications of biogas upgrading in the country are given in detail.

3. Biogas Valorisation to Biomethane

The techniques for biogas upgrading can be classified into sorption, separation, and in situ techniques, as shown in Figure 3. Sorption and separation are further classified depending on the basis of operation: adsorption, physical or chemical absorption, high- or low-pressure permeation/membrane separation, and cryogenic methods [7,19,20]. Each method comes with its advantages and disadvantages, such as social-economic effects, ambient impacts, methane loss, amount of energy needed for the process, complexity of using the equipment, and the accessibility of equipment or skilled workforce to operate the equipment [8,21,22]. In situ techniques involve biological methods that make use of living organisms to convert CO_2 to CH_4 and H_2O inside the biodigester. This results in an increase in CH_4 from 60% to 96%, together with a reduction of H_2S to insignificant levels. Various chemicals are also used to convert CO_2 to CH_4 in biodigesters.

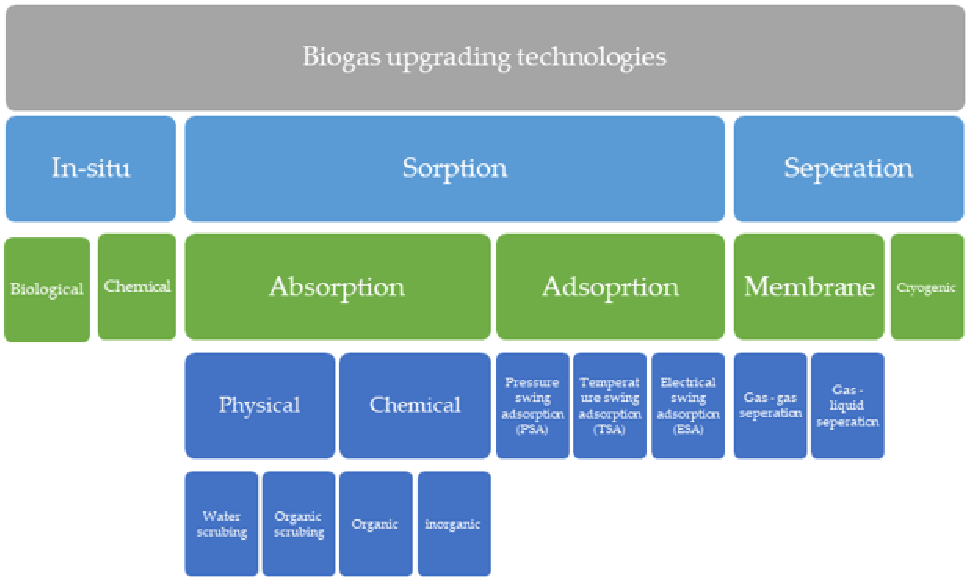

Figure 3. Biogas upgrading techniques, adapted from [7,19,20].

3.1. Common Methods of Biogas Upgrading to Biomethane

Various techniques have been discussed in depth by a number of researchers [7,8,10,20,21,23–26]. Characteristics of the commonly used upgrading/cleaning techniques are given in Table 2. Absorption is divided into physical (water and organic) and chemical scrubbing. Water scrubbing makes use of higher solubility of CO_2 and H_2S than the CH_4 in water. For example, CO_2 is 26 times more soluble than CH_4 at 25 °C. Organic scrubbing is similar to water scrubbing, in that it makes use of a different solubility, but it requires higher energy at higher temperatures for the regeneration of solvents. Popular solvents are selexol®, rectisol, and genosorb®, and they can absorb CO_2, H_2S, and H_2O.

Table 2. Characteristics of common biogas upgrading techniques.

Parameter Method	Water Scrubbing	Organic Scrubbing	Chemical Absorption	Pressure Swing Adsorption	Membrane Separation	Cryogenic Separation
Basis of operation	Physical absorption	Physical absorption	Chemical absorption	Adsorption	Gas—gas Permeation Gas—liquid Absorption	Multistage compression and condensation
Temperature (°C)	20–40	10–80	35–50	5–30	25–60	−110–−25
Biogas pre-treatment requirement	H_2S removal recommended to avoid its release into the atmosphere from the desorption tank	Removal of H_2S	Removal of H_2S	Use activated carbon to remove H_2S and dry to remove H_2O	Use of sodium hydroxide, polyvinyl alcohol, and H_2O to remove H_2O and H_2S	Compress biogas to 17–26 bar and cool to −26 °C to remove H_2S, H_2O, halogens, and siloxanes
Working pressure (MPa)	0.4–1.2	0.4–0.9	0.1–0.2	0.4–1	2–3.6 high pressure systems 0.5–0.8 low pressure systems	1–8
Solvent/adsorbent/ consumables	H_2O_l, antifouling, and drying agents	Organic solvents, polyethylene glycol	Amines, alkalines, antifouling, and drying agents	Adsorbents, molecular sieves	Selective membranes	Glycol refrigerant
Heat requirement	No	Intermediate	High	No	No	No
Energy consumption ($kWhNm^{-3}$ biomethane)	0.2–0.5	0.10–0.67	0.05–0.27	0.16–0.46	0.18–0.43	0.2–0.79
CH_4 purity (%)	95–99	95–99	>99.0	95–99	95–99	96–99
Methane loss (%)	1–10	1.5–4	0.04–0.1	1–4	0.5–0.6	0.5–3
Capital cost (€Nm^{-3} biogas)	0.13–15	0.25	0.28	0.26	0.5–20	0.5–3
Running costs (€Nm^{-3} biomethane y^{-1})	0.091	0.090	0.112	0.092	0.065–0.101	
Easiness of operation	Simple	Difficult	Difficult	Extensive monitoring required	Easy	Complex
Technical availability (%)	96	96	91	94	98	-

Chemical scrubbing is based on dissolving impurities, such as CO_2 and H_2S, in biogas via a reversible chemical reaction between the absorbent and the absorbate. Commonly used absorbents, which dissolve more gases per unit solvent volume than water, include monoethanolamine (MEA), diethanolamine (DEA), and triethanolamine (TEA). Thus, smaller upgrading units are required in comparison to water scrubbing units. However, the requirements for absorbent regeneration make the process more expensive than water scrubbing.

Pressure swing adsorption (PSA) is a mass transfer process, in which the adsorbent selectively retains some molecules of biogas on its surface, based on molecular size, which is driven by differential partial pressure. For example, an adsorbent material with 0.37 nm pores will retain CO_2, which has 0.34 nm pores, while allowing CH_4, which has pores that are 0.38 nm. Although H_2S is retained, it is advisable to pre-treat the biogas to remove H_2S because it makes it difficult to regenerate the adsorbent. The effectiveness of the adsorption process is governed by the pressure of the adsorbate, the temperature of the system, and the size of the pores of adsorbent material. Common adsorbents include activated carbon, metal-organic frameworks, zeolites, silica gel, and molecular sieves. To reduce CH_4 loss, Augelletti et al. (2016) proposed and simulated an adsorbent process that utilised Zeolite 5A adsorbent material in two sections [27]. The biogas is input into the first section, where 98.9% pure CH_4 is produced. Methane recovery in this section is low; thus, the off gas from this section is passed onto the second section, which removes CO_2 and returns the gas to the first section for methane capture. This process results in methane recovery of more than 99% while consuming approximately 1.25 $MJkg^{-1}$ of biomethane. Thus, there is great potential for the improvement of the PSA technology performance.

Membrane separation is based on the selective permeation of certain gas molecules through a selective permeable membrane. The biogas separates into the permeate fraction, comprised of CO_2, H_2O, O_2, and H_2S, and the retentate fraction, made up of CH_4 and N_2. The most-used membranes are polymeric membranes, which are made from organic materials, such as polysulphone, polyimide, polycarbonate, polydimethylsiloxane, and cellulose acetate. The less-popular membranes are inorganic and mixed matrix membranes.

The cryogenic process separates CH_4 from the impurities in biogas using difference in phase change temperature and distillation characteristics of the constituent molecules. At 1 atm, CO_2 condenses at −78.2 °C and is removed from biogas, leaving mainly CH_4, which has a boiling point of −161.5 °C. To prevent the clogging of pipes from frozen substances, H_2S, H_2O, siloxanes, and halogens are removed before cryogenic cleaning. This technology is still in its infancy but has started to penetrate the market. The simplest cryogenic process maintains a constant pressure of 10 bar, while the temperature is decreased in steps that remove the impurities. At −25 °C, H_2O, H_2S, and siloxanes are removed. The second step partially removes CO_2 at −55 °C by liquefication, while the last step removes all CO_2 at −85 °C by solidification.

The conventional technologies described above need pre-treatment to remove mainly H_2S, as shown in Table 2. The technologies that remove both CO_2 and H_2S have higher running costs and significant negative impacts on the environment. Microalgae-based technologies have been developed to remove both CO_2 and H_2S from biogas [28–31]. These processes use CO_2 for microalgae photosynthesis and use the oxygen produced during photosynthesis to oxidise H_2S. Several experiments were carried out with a high-rate algal pond (HRAP) connected to an external absorption column (AC) for the simultaneous removal of CO_2 and H_2S [29,32,33]. Although there was the 100% removal of H_2S, that for CO_2 was not 100%, and in some cases, it was less than 80%. Additionally, the produced gas contained nitrogen and oxygen gases, which presents a drawback of this technology in upgrading biogas. To overcome this hinge, Toledo-Cervantes et al. (2016) optimised both the process of the photosynthetic upgrading of biogas and the recovery of nutrients from the digestate in an algal-bacterial HRAP that is connected to a biogas AC by recirculating the settled liquid [28]. This reduced the concentration of CO_2 to 0.4 ± 0.1%, oxygen to 0.03 ± 0.04%, and nitrogen to 2.4 ± 0.2%, while increasing the CH_4 purity to 97.2 ± 0.2%, which meets most European biomethane standards.

Another upcoming technology that simultaneously removes CO_2, nitrous oxides, and H_2S from biogas is the adsorption of impurity molecules on a solid surface, making use of physical or weak van der Waals forces that cause different gaseous molecules to selectively attach onto solid surfaces [34]. Porous materials, such as biochar, clay, and silica gel, are used as adsorbents. Mulu et al. (2021) compared clay dry adsorption with wet carbonation methods of upgrading biogas [35]. For wet carbonation, the optimal clay-to-water ratio was 1:3 at 75 °C. Sodium hydroxide-activated clay increased CO_2 by more than five times. H_2S was 100% removed, and removal rate of 93.8% for CO_2 was achieved. Sethupathi et al.

found that the adsorption capacities of biochar differ according to its properties [36]. The successful use of clay and biochar is beneficial to small-scale digesters, who find the conventional technologies to be expensive because of low economies of scale [7]. A novel process involving the use of the dry adsorption of wood ash and carbonation technologies in the upgrading of biogas was created [37]. In this study, 88% CH_4 purity and 2.30 mmolg^{-1}-wood ash CO_2 uptake were achieved. A biogas flow rate of 100 mlmin^{-1} was achieved at an adsorbent-to-water ratio of 1:4 for carbonation. Changing the mass of activated wood ash in dry adsorption from 2.5 to 35 g resulted in an 8.9 to 67.9% increase of CO_2 removal, which is not a significant improvement. Thus, it is advisable to use raw wood ash for household AD.

Some researchers proposed that ex situ biogas upgrading technologies are economically viable when used on a biogas plant that produces more than 2400 m^{3}d^{-1} biogas and recommended in situ for small/medium biogas plants [38]. There are a number of in situ methods, such as the addition of H_2 into the reactor, bio-electrochemical systems, high-pressure anaerobic digestion, and the addition of additives such as biochar and an up-flow anaerobic sludge blanket.

The technologies (first generation biogas upgrading technologies) described above have the disadvantage of not utilising CO_2 as an energy source. Researchers have started working on second generation biogas upgrading technologies, which make use of CO_2 for energy storage [39]. Most of these strategies are still in the demonstration stage. In Denmark, a demonstration plant was set to catalytically convert the CO_2 from biogas to CH_4 [40,41]. The first section of the plant removes most impurities from biogas and converts them to nutrients that can be added to slurry. The biogas is then passed on to the second section, where CO_2 reacts with green H_2 to produce CH_4 using the Sabatier process. The green hydrogen is produced from the electrolysis of water using wind energy. A stable steady-state CO_2 conversion rate was achieved, with more than 90% of CO_2 turned to biomethane.

3.2. Biogas Industry Status in South Africa

AD in South Africa started by the construction of a digester at a pig farm in Johannesburg in 1957, and the gas was used to run a six-horse-power Lister engine [16,42]. Since then, biodigesters have been installed by households, farmers, various institutions, and companies. There has been growing effort to understand opportunities and challenges in the biogas industry. This has led individuals, companies, government departments, and researchers to form organisations that work on biogas; one of these is the Southern African Biogas Industry Association (SABIA) in 2013, which has the representation of the industry in the country. In 2021, SABIA and the World Biogas Association requested the South African Minister of Environment, Forestry, and Fisheries to integrate biogas into national plans to enhance the achievement of the government commitment to the Paris Agreement [43]. This would enhance the growth of the biogas industry. Currently, 43 organisations, including one academic institution, are registered with SABIA [44]. Most of these companies have websites, but not much information is found on some of them.

Prior to 2000, mainly small-scale biodigesters were installed to produce biogas for space heating and cooking from livestock manure [42]. To date, the feedstock is diverse, including slaughterhouse waste, chicken waste, fruit waste, sugarcane bagasse, municipal solid waste, wastewater, cattle waste, and pig waste to name a few [16]. For the feedstock sources captured in the national biomass inventory, of which the majority is sugar and municipal solid waste in the KwaZulu-Natal, Gauteng, and Western Cape Provinces, it has been estimated that a total of 3 million Nm^{3}d^{-1} of biogas can be produced, as shown in Figure 4 [45]. Awareness raising and information sharing are being promoted, and tools that will enhance decision-making by interested stakeholders, such as a biogas guidebook for South Africa, have been developed, and some are available for free on websites [46].

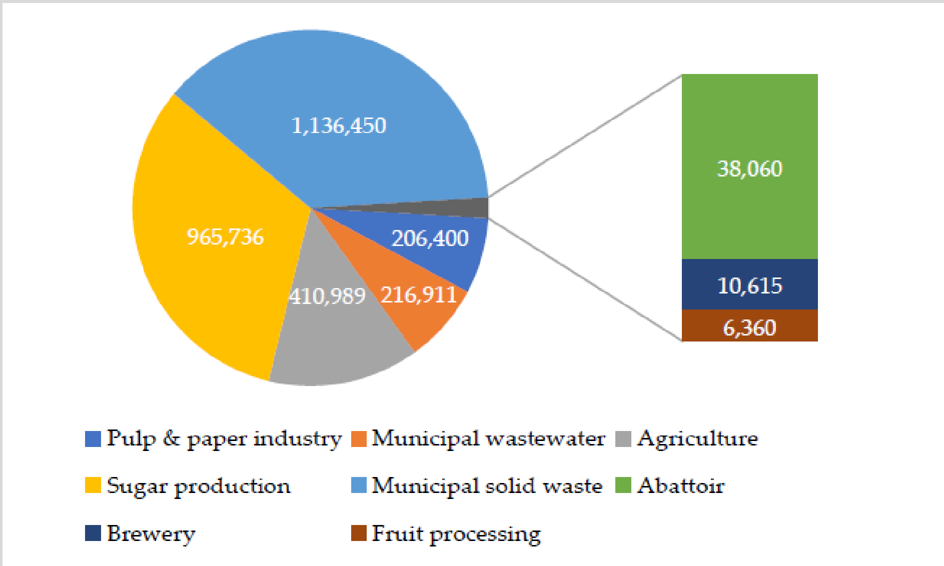

Figure 4. Biogas potential yield by sector (Nm^3d^{-1}) [45].

The number and size of biogas plants have increased since the introduction of the technology, with household/domestic/micro producing <0.5 kW_e, small-medium (0.05 $\geq$ 1 MW_e) and large-scale digesters being installed across the country [18,47]. In 2018, a total of 700 biodigesters were installed [16,47]. As a result of difficulties in finding information on the number of installed biodigesters, the 2018 installation of 350/700 was used to set a baseline for micro-biodigesters at 350 biodigesters in 2021 [47,48]. Thus, this research assumed the same baseline year and total installed biodigesters. This implies that in 2021 there were 350 small-medium and large-scale biodigesters. Table 3 gives an overview of the installed small-medium and large-scale biodigesters. Plant benefits include selling electricity, saving by not buying electricity from Eskom, collecting gate fee revenue, and selling CH_4 and CO_2. In addition to these monetary benefits, the facilities also practice good waste management, which keeps their environments clean.

Table 3. An overview of the installed small-medium and large-scale biodigesters.

Location	Substrates	Biogas Production (Nm^3h^{-1})	Power Output (MW)	Revenue Source
Bronkhorstspruit	Agricultural and agricultural processing waste		4.6	Electricity, gate fees
Grabouw	57 $tond^{-1}$ food and abattoir waste		0.527 and 0.55 MW_{th}	Electricity and heat cost savings
Darling	Bovine manure		0.5	95% electricity cost savings
Bronkhorstspruit	Farm residue	60	0.1	Electricity savings
Athlone,	Municipal solid waste	1200		CH_4, CO_2, gate fees,
Durbanville	35–45 m^3d^{-1} Pig waste	22	0.075 and 0.1 MW_{th}	Heat and electricity cost savings

Source [16,18,49,50].

Currently, most biogas is being converted into electricity. A financial analysis was performed for small-medium scale commercial biogas plants that convert biogas into electricity under the current scenario, where waste is dumped in landfills and energy cost is low with subsidies from the government [18]. Small-scale plants were found not viable, while medium-scale plants were viable for facilities that have high waste management costs, such as abattoirs. The conclusions made indicated that high waste management costs, the availability of huge feedstock volumes, and high on-site energy needs enhance the financial viability of biogas commercialisation.

The price of electricity on the market is half the price of compressed biomethane when both are measured in gigajoules, being R235 GJ^{-1} and R111 GJ^{-1} for transport fuel and electricity, respectively [51,52]. Additionally, the taxes and levies that are put on liquid fuels are exempted on gaseous transport fuels. This makes the use of biogas as a fuel a more attractive option on the market. Thus, biogas producers will want to sell their gas to the transport sector if the infrastructure permits. However, their hopes may be frustrated by limited gas-filling stations, which are still very scarcely scattered in the country, as shown in Figure 5. Taking into account the limited gas-fuelling stations and the limited range travelled by a full tank of gas vehicle compared to conventional liquid fuel (diesel or petrol) vehicles, it is recommended that the focus be on fuelling vehicles that have recurring short routes, such as minibus taxis and public city buses [45].

Figure 5. The location of natural gas fuelling stations [53].

An assessment used to find the feasibility of using biomethane as a fuel for public transport revealed that a total of 230 $ktony^{-1}$ dry feedstock is available in Johannesburg and can generate 91.6 million Nm^3y^{-1} fuel grade CH_4, which can be used by approximately 2700 buses [54]. The study identified two sites that can be used for locating more than 2000 Nm^3h^{-1} biogas facilities to produce biogas and upgrade it to fuel up to 600 city buses [52].

The biogas sector is faced with historical challenges that may hinder the uptake of the technology, and as a result, all efforts used to valorise biogas into biomethane will be in vain. They include high capital costs for the design and construction of biogas digesters; the seasonal variation of feedstock, which may result in failure of the digester because of insufficient substrates; limited research to come up with local solutions to operational challenges; the cheap disposal of waste to landfills; the failure of previously installed biodigesters; and the presence of impurities in biogas, which reduce its calorific value and damage appliances using the gas [17]. Another drawback is the absence of a long-term demand for biomethane and enabling regulatory framework to support investment in biogas upgrading [45].

There are opportunities in the sector that would enhance its growth. The Renewable Energy Independent Power Producer Procurement Program (REIPPPP) aims to produce electricity from renewable energy resources, such as wind and biomass [55]. SABIA encourages municipalities and Eskom to buy surplus electricity from biogas plant operators [56].

The National Environmental Management: Waste Act has resulted in an improved regulatory environment; however, it still needs further improvement [57].

3.3. Biogas Upgrading Technologies Tried in South Africa

Not much work has been published on biogas upgrading activities by companies in the country. One company uses a combination of an advanced membrane and a cryogenic upgrading system to upgrade biogas. It appears that websites are created and work is published when there are funded projects [58,59]. At the end of the project, the websites are not updated. Therefore, the continuity of the activities is uncertain. However, on the research side, there is evidence of work going on in the area of biogas upgrading. If researchers collaborate with industries, more work can be published, and this would enhance information dissemination to benefit various stakeholders, who will learn from the experiences of others.

A group of researchers have conducted several experiments and simulations on biogas upgrading techniques. Their research started with a detailed review on biogas upgrading techniques that are used to obtain vehicle grade fuel and opportunities and challenges in its market [60,61]. Their review and the subsequent research work showed that the country has people who understand these technologies. Hence, the uptake of such technologies can be facilitated by the collaboration of researchers and the industry.

An experimental investigation was performed to find the effectiveness of the organic solvent, monoethanolamine (MEA), and inorganic solvents, sodium hydroxide (NaOH) and potassium hydroxide (KOH), in producing biomethane from biogas produced from the co-digestion of various feedstock [62–64]. The feedstock included cow dung, chicken droppings, and fruit wastes from a farm and bins in the Johannesburg city commercial market. The chemical absorbents absorbed CO_2, H_2S, and sulphur oxides (SO_x) from biogas, resulting in an increased CH_4 quantity in the biogas. MEA and NaOH were found to be more effective than KOH in cleaning the biogas to produce biomethane because they have higher absorptive capacities. It was shown that increasing the concentration of the absorbent increased the rate of absorption and the impurity removal efficiency by the absorbent.

Biomagnetic nanoparticles (BMNPs) copper (II) oxide (CuO), iron (III) oxide (Fe_2O_3), titanium (IV) oxide (TiO_2), and a combination of Cu/Fe-TiO_2 were used as bio-stimulants for lab-scale wastewater treatment and biogas upgrading to more than 90% CH_4 [65]. The wastewater and sludge were obtained from a sugar refinery plant in KwaZulu Natal (KZN) Province in South Africa. The AD process was carried out at a mesophilic range, with a hydraulic retention time of 30 days. The use of BMNPs significantly improved the biogas production and CH_4 purity, which increased from 63% to 100%. The wastewater treatability performance (chemical oxygen demand, colour, turbidity, total suspended solids, and volatile solid removal) increased from 45–65% to 75–90%. The morphology and elemental distribution of the BMNPs and their biomagnetic interactions with the microbes facilitated the adsorptive removal of the toxins and the biogas yield, with best result coming from the Cu/Fe-TiO_2 addition. Bio augmentation using BMNPs proved to be an economically viable method for effective organic waste management to produce biogas of a high CH_4 purity.

Amo-Duodu et al. (2022) used bio-chemical methane potential (BMP) tests to investigate the effects that the ferrites aluminium ($AlFe_2O_4$) and magnesium ($MgFe_2O_4$) magnetic nanoparticles (MNPs) had on biogas production [66]. Three reactors were used. The first one was the control, containing 500 mL of wastewater and 300 mL of activated sludge. The other two were comprised of 1.5 g of the two MNPs, which were each added to a mixture similar to the control. The BMPs increased chemical oxygen demand (COD) in comparison to the control. $MgFe_2O_4$ had 93.96% COD degradation, while $AlFe_2O_4$ and the control had 85.95% and 68.83%, respectively. $MgFe_2O_4$ produced biogas of 85.7% CH_4 purity, and it was 84.3% and 65.7% for $AlFe_2O_4$ and the control, respectively. Although the MNPs performed better than the control, there is still need for improving their performance through more experiments.

Titanium dioxide magnetite (Fe-TiO_2) was applied to a mixture of wastewater and inoculum (anaerobic sewage sludge) from the KZN Province sugar refinery plant in a lab-scale reactor at 35 °C for 30 days [67]. The results were compared with the control reactor, where AD occurred without a nanoparticle stimulant, and another reactor, where AD was stimulated by Fe-TiO_2 additives in a magnetic field. The AD + Fe-TiO_2 and AD + Fe-TiO_2 + magnetic field reactors produced biogas with 100% methane concentration, while the AD reactor produced biogas with a 66% CH_4 concentration. The induced magnetic field was found to be an environmentally friendly method for treating wastewater to produce CH_4-enriched biogas. The Fe-TiO_2-augmented AD reactor in a magnetic field had the highest treatability and biogas production of 30 mLd^{-1} in comparison to 20 mLd^{-1} for the control and 25 mLd^{-1} for Fe-TiO_2 + AD reactors. Over a period of 30 days, the total gas production was 815 mL for the Fe-TiO_2-augmented AD reactor in a magnetic field, 680 mL for the Fe-TiO_2 + AD reactors, and 625 mL for the AD-only reactor.

Tetteh et al. (2022) stimulated the AD of wastewater using Fe-TiO_2 coupled to ultraviolet light, thus forming a bio photo-reactor (BP) [68]. To investigate the effect of the BP and the bio photo-magnetic (BPM) on AD, they used three experimental setups at 35 ± 5 °C with a 30-day retention period; the control was made up of an AD system without a catalyst, a BP system, and a BPM system, made up of the BP system in a magnetic field. The BPM system produced 1.33 Ld^{-1} biogas of 98% CH_4 purity, with 75% COD removal at a pH of 6.5. The BP and the control produced 1.125 Ld^{-1} and 0.625 Ld^{-1} biogas, respectively, with corresponding 65% and 45% COD. The AD system biogas contained 65.5% CH_4. Coupled with a CO_2 reduction of 0.64 kg CO_2L^{-1}, the use of BPM is a promising technology for biogas production and in situ upgrading. In a separate experiment, the BPM system showed more than an 85% decrease in colour, COD, and turbidity [69]. The emission of CO_2 to the environment was reduced by 1.74 kg CO_2m^3.

Lottering (2019) suggested the use of hybrid biogas upgrading techniques to obtain high CH_4 yields at low cost and low energy consumption [22]. Various feedstocks were used in lab experiment to evaluate and optimize a plant for upgrading biogas. The biogas was produced at mesophilic range and atmospheric pressure. The plant utilised a combination of water scrubbing and membrane separation to produce a ±98% CH_4 purity. CHEMCAD was used for the size and cost of the plant, which would attain the gas Wobbe index, relative density, and CO_2 and oxygen levels required by German standards. The production cost was found to be R21 kg^{-1} and R30 kg^{-1} for CH_4 and CO_2, respectively. The payback period was found to be 5.65 years.

Biochar was produced from the pyrolysis of biomass, and the investigation was carried out to find its effect on mesophilic anaerobic digestion and the in situ upgrading of biogas to approach a state of biomethane [70]. A Taguchi-based design batch experiment was carried out to test the biochemical CH_4 in the lab. The parameters that were investigated included the mixing ratio of fruit and vegetable waste and kitchen waste and the effect of biochar quantity, production temperature, and particle size on the performance of the AD. The performance of the AD was measured in terms of specific biomethane, percentage concentration CH_4, the electrical conductivity, and pH. The optimal conditions that were obtained from the batch experiment were repeated in a semi-continuous two-stage experiment in a biochar-modified digester and a control digester. A techno-economic analysis was conducted for biochar-modified AD. The concentration of CH_4 in the biochar-modified biodigester was in the range of 73.38–77.61%. This was 19.24% higher than that of the control digester. Both the CH_4 and economic analysis of the biochar-modified digester did not justify investment into the biochar-modified AD at a mesophilic range.

Masebinu et al. (2014) simulated the use of gas permeation technology in upgrading biogas to CH_4 using ChemCAD [71,72]. A double-stage membrane and single-stage configuration were considered to compare the effect of recycling the permeate stream. The membrane section was first simulated for biogas enrichment only and then for combined cleaning and enrichment. The results showed the 100% removal of H_2O, and H_2S. 18% improvement on CH_4 recovery was achieved by recycling the permeate stream to an overall 81.23% CH_4 recovery. Various parameters were found to affect CH_4 recovery, which decreased by 4.1% with an increase in the amount of CO_2 in the feed from 10 to 60% and increased with pressure increase [73]. At the same time, CH_4 recovery increased by 7.11%, while the membrane separation area decreased by 47.73%, with a 5.6 to 33.33 increase in membrane selectivity. An 80–140 m^3h^{-1} gas flow rate increase resulted in a 6.2% CH_4 recovery and 2.6% membrane area increase. CH_4 purity and recovery were found to be 91% and 96%, respectively, at optimum operating conditions, with a gas processing cost (GPC) of USD 0.36 Nm^{-3}.

From the products available on biogas companies' websites, which have upgrading equipment, it can be implied that membrane separation and PSA are being used in the country [74,75]. If researchers are working with companies who are using the technologies, it can be deduced that chemical absorption and in situ upgrading using BNPs are being used.

However, most conventional biogas-upgrading technologies are mature or near mature and are available on the markets and, hence, can be used locally. The advantages and disadvantages of these technologies are well documented in most literature [7,8,12]. Water scrubbing can be easily operated, removes CO_2 together with H_2S, and has low capital cost, but its high water demand, damage of equipment by H_2S, and possibility of growth of bacteria, which causes the blockage of equipment, make the technology less desirable for industrial applications. It can be used for domestic purposes where technical expertise might not be available all the time. The difficulties in organic scrubbing operations make it ideal for industrial applications, where necessary expertise can be always available. Chemical scrubbing is recommended for plants that supply biomethane to companies that need more than 99% methane purity because it is difficult to operate and has high operational costs. PSA requires extensive process monitoring, which can be performed remotely, but is relatively inexpensive; therefore, it is more recommended than chemical scrubbing for uses where % methane purity is not of high priority. Although membrane separation is easy to operate and has low running costs, it may have relatively high capital costs.

3.3.1. Cost and Energy Implication of Biogas Beneficiation Methods

The use of biomethane to replace fossil fuels will result in primary energy security. Life cycle cost (LCC) assessments are usually used to compare the benefits of biogas-upgrading techniques in terms of environmental, energy, and economic impacts [76,77]. This analysis gives monetary value to the environmental impacts of the whole life cycle for biomethane, from production to the end-of life management, in addition to the energy and economic monetary vales. LCC assessments conducted for membrane separation, water scrubbing, chemical scrubbing, and PSA varied between −203 EUR/FU for PSA and −210 EUR/FU for membrane separation. This indicated that the revenue from biomethane and savings from avoided fossil fuels are greater than the direct and indirect costs of producing biomethane [77]. The energy requirements, initial investment, and operational costs for the commonly used technologies are given in Table 2.

The energy value of 1 Nm^3 gaseous fuels is given in Table 4. Biomethane is shown to be competitive with fossil fuels; thus, it can be used as a substitute for fossil fuels in all their applications [8]. Petrol is equivalent to 1.1 L and 1.2 L of 1 m^3 biomethane and natural gas, respectively. Approximately 20% of the energy in biogas is used for upgrading it to biomethane, and no external energy is required, making the process self-sustaining. The cost of this energy is 77.4%, 85.4%, 60.1%, and 66.3% of the operating costs for membrane separation, water scrubbing, chemical scrubbing, and PSA, respectively [77].

Masebinu et al. (2015) used ChemCAD to simulate the economic performance and sensitivity analysis of a privately owned biogas-upgrading plant in Sebenza, South Africa [78,79]. The plant makes use of a hollow fibre membrane to separate CO_2 from the raw biogas to attain the preferred fuel grade CH_4. The cost of processing the gas (CPG) was found to increase with increases in CO_2 concentration and the pressure of the feed. The increase in the flow rate of the feed increased the flow rate of the product and decreased the CPG. The net present value, the benefit cost ratio, the internal rate of return, and the payback period were found to be R15 240 343, 2.05, 22.41%, and 5 years, respectively. The study revealed that savings on yearly fuel costs of up to 34% could be made if gasoline is replaced with compressed CH_4 and the payback period for retrofitting the vehicle is 1.25 years. Table 5 gives an analysis of profits made by an end user when using compressed biomethane gas, petrol, or compressed natural gas for mini-bus taxi fuel. The results of this study implied that the use of compressed CH_4 as fuel for vehicles is the most cost-effective option for the compared fuels.

Table 4. Energy value of different fuels.

Fuel	Energy Value (MJ)
60% CH_4 biogas	21.6
97% CH_4 biomethane	34.812
Natural gas	39.6
Petrol (1 L)	32.616
Diesel (1 L)	35.28

Adapted from [8].

Table 5. Profitability of using biomethane as vehicle fuel in comparison to other fuels.

Parameter Compared	Petrol	Natural Gas	Biometahane
Yearly fuel consumption (L)	9770	9770	9770
Yearly fuel cost (R)	138,153.35	97,606.22	90,897.95
Yearly monetory savings on fuel consumption (R)		40,547.13	47,255.40
Payback period (years)		1.5	1.3
Savings (%)		29	34

Adapted from [78].

Membrane separation, water scrubbing, chemical scrubbing, and PSA are mature technologies that are used mainly in developed countries to produce biomethane that can be used to substitute fossil fuels in all their applications and use natural gas pipelines and LPG bottles for transportation and storage. These methods have small variations in terms of capital costs ranging from 0.13 €Nm^{-3} biogas to 3 €Nm^{-3} biogas for water scrubbing and cryogenic separation, respectively, given in Table 2. Additionally, the running costs show little variation. All these technologies have negative LCC, which implies that the monetary benefits of using them are greater than the biomethane production costs.

3.3.2. National/Environmental Significance of the Beneficiation of Biogas

The presence of impurities in biogas that damage appliances that transport, store, or use biogas lead to the rejection of biogas technologies [80]. This negative attitude is changing as a result of the introduction of methods for cleaning the gas to remove the toxins, which lead to wider applications of biogas. It has been proven that upgrading biogas into biomethane results in a clean gas that can substitute for fossil fuels, such as natural gas, petrol, and diesel in the transport sector. The valorisation of biogas has the following benefits: improved waste management, which avoids landfills and its associated CH_4 emission; employment creations; increased access to affordable energy; and increased energy security.

The benefits of producing and upgrading biogas to biomethane can be clearly illustrated using Figure 6, which shows the production and beneficiation process at a facility plant in Athlone, Cape Town. This state-of-the-art biogas plant was set up with an initial investment of around R400-million and was commissioned in 2017 [18,46]. The plant created 80 permanent jobs and hundreds of indirect jobs. Thus, the surrounding communities' livelihoods can be improved by earning a steady income from employment. After waste separation, 200–300 $tond^{-1}$ organic/biodegradable waste is fed into the biodigester. This waste is diverted from landfills, thus preventing emissions of GHG into the atmosphere. A total of 1200 Nm^3 of biogas is produced per hour. It is upgraded using a combination of advanced membrane and cryogenic upgrading systems to produce approximately 760 Nm^3 of 95.5% purity CH_4 and 740 Nm^3 of liquid CO_2 [15,18]. The CH_4 is compressed and bottled in already existing LPG gas cylinders and used as a substitute for wood or fossil fuels in household cooking and the transport industry, respectively. This reduces deforestation and increases energy security. The CO_2 is upgraded to food-grade CO_2 and transported to industries for various uses. This further reduces the emission of GHG into the atmosphere. If other similar plants are installed in the country, it will lead to improved livelihoods of many people, energy security, and increased contribution to the abatement of climate change. Thus, it is important for researchers to partner with industrialists and conduct detailed techno-economics for the implementation of a new biomethane, generating units on the scale that exists in Cape Town.

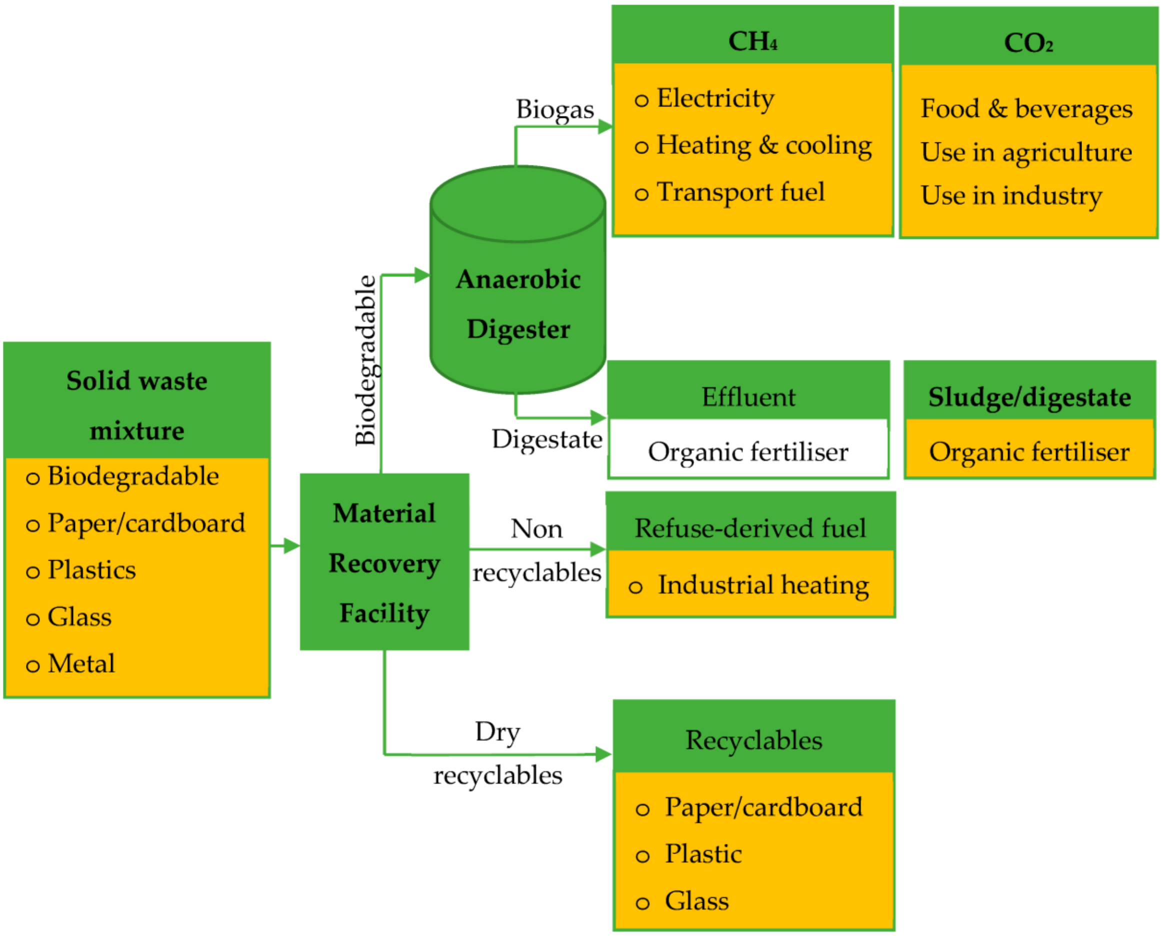

Figure 6. Benefits of biogas production and the beneficiation system.

Biomethane can be used a substitute for natural gas in all industrial applications if upgraded enough to meet the required quality standards. Already, Sasol produces wax, jet fuel, diesel, and petrol via the Fischer-Tropsch process using natural gas; hence, it can displace natural gas with biomethane. Table 6 shows that biomethane has the lowest impact on the environment in terms of the entire lifecycle (well-to-wheel) of GHG emissions in comparison to popularly used fossil fuels [23]. Thus, it is an environmentally friendly substitute for fossil fuels in the transport sector, where the GHG emission savings resulting from avoided fossil fuel reach 90% [46]. Since 2014, natural gas-filling stations for vehicles have been operational in South Africa, and biomethane usage can make use of these already existing facilities.

Table 6. Comparison of well-to-wheel (W2W) GHG emissions of different fuels.

Fuel Type	W2W Emissions gCO_2eqkm^{-1}
Petrol	164
Diesel	156
Compressed natural gas (CNG)	124
Compressed biomethane (bioCNG)	5

Source [23].

CO_2 is the major by-product of biogas upgrading, and capturing it will reduce the emission of GHG to the atmosphere. CO_2 is usually vented into the atmosphere or it is captured, compressed, and transported using carbon capture and sequestration techniques and injected into carbon sinks, such as rock formations [19,81]. The facility in Athlone upgrades CO_2 into food-grade CO_2, which is compressed, bottled, and used in the food and beverages industry [18]. Thus, this further reduces the amount of CO_2 released into the atmosphere.

Feasibility studies on the use of biogas as a fuel for city buses have shown that more than 7 ha of land is required to grow energy crops to produce biofuel to be used by one bus [52,54]. Thus, the use of biomethane for the fuel sector avoids competition for land with food and feed production. In addition, clean environments will be created by diverting biodegradable waste from landfills, avoiding particle matter emissions of 30 $tony^{-1}$, and reducing nitrogen oxide emissions by 1.6 $tony^{-1}$ by fueling only 600 buses. Thus, significant environmental benefits are achieved if biomethane is used as transport fuel for the whole country, starting with public transport in selected cities that have gas-fueling service stations.

4. Discussions

The current review used work that was published in the last 10 years about biogas-upgrading techniques that are used globally to increase the biogas calorific value and make it suitable for automotive fuel or for use in conventional natural gas applications. More focus was given to South African-based activities. The literature was obtained from various online databases and websites. The advantages of sorption, separation, and in situ technologies were discussed.

The biogas industry was described in detail. SABIA was established in 2013 for the biogas sector to be represented on the national and international fora. A total of 43 organisations are currently subscribed to SABIA. The KwaZulu-Natal, Gauteng, and Western Cape Provinces provide the bulk of the anticipated 3 million Nm^3d^{-1} biogas to be produced in the country. The future development of the biogas sector will be compared to a set baseline of 350 micro biodigesters and 350 small-medium and large-scale biodigesters in 2021. Benefits of biogas facilities include the selling of electricity, saving money by avoiding purchasing electricity from Eskom, collecting gate fees, and the selling of CH_4 and CO_2 depending on biogas usage. Upgrading biogas to biomethane is more profitable than converting it to electricity. REIPPPP can be exploited by entrepreneurs to develop biogas plants. However, there is no roadmap for the production and usage of biomethane in the short- and long-run.

On the research front, several activities have been recorded. Laboratory work, field work, simulations, and article reviews have been published. The use of BMNPs in bio augmentation was found to be a financially viable strategy for managing bio-degradable wastes and the production of biogas with a high CH_4 purity. The use of biochar and MNPs did not perform well in AD systems. From information gathered from websites, it can be inferred that membrane separation and PSA are being used in the country. Additionally, some websites give the indication that a combination of advanced membrane and cryogenic upgrading systems are being used. Outputs from research activities imply that chemical absorption and in situ upgrading using BNPs are also being used.

Biogas upgrading facilities are self-sustaining in terms of energy. For facilities that are operational in the country, the heat and electrical energy is used to meet the company's energy requirements, and excess is sold mainly to municipalities. The upgrading of biogas by the removal of CO_2, to form fuel-grade CH_4 using hollow fibre membranes and the retrofitting of the vehicle to use the biomethane were found to be cost effective. The payback periods were 5 years and 1.25 years for the upgrading system and the retrofitting of the vehicle, respectively.

Some facilities compress and bottle the biomethane in already existing LPG gas cylinders for the replacement of wood or fossil fuels in household cooking and the transport industry, respectively. More value is created by upgrading the CO_2 to food-grade CO_2 for industrial use. Thus, the use of both CH_4 and CO_2 results in reduced deforestation, increased energy security, and further reduction of the emissions of GHG into the atmosphere. This diversification of biogas usage results in more jobs being created than those in facilities that produce electricity from biogas.

5. Final Considerations

The pros and cons of conventional biogas upgrading technologies that are available on the market are well known. H_2O scrubbing has several disadvantages, which make it unsuitable for industrial applications. However, it is recommended for micro-scale usage because of its easiness to operate in the absence of skilled manpower to manage the systems. For facilities whose customers require more than 99% CH_4 purity, this study recommends the use of chemical scrubbing. PSA can be used instead of chemical scrubbing by facilities whose customers are not concerned with high CH_4 purity. Although membrane separation is easy to operate and has low running costs, it may have relatively high capital cost and is thus recommended for large-scale plants where economies of scale would increase the chances of profitability.

Researchers in academic institutions and research centres are recommended to collaborate with the industry and entrepreneurs so they can work together to adapt the biogas-upgrading technologies to the local environment.

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