REVIEW

Integration of biogas systems into a carbon zero and hydrogen economy: a review

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Abstract

The Ukraine conflict has put critical pressure on gas supplies and increased the price of fertilisers. As a consequence, biogas has gained remarkable attention as a local source of both gas for energy and biofertiliser for agriculture. Moreover, climate change-related damage incentivises all sectors to decarbonise and integrate sustainable practices. For instance, anaerobic digestion allows decarbonisation and optimal waste management. Incorporating a biogas system in each country would limit global warming to 2 °C. If suitable policies mechanisms are implemented, the biogas industry could reduce global greenhouse gas emissions by 3.29–4.36 gigatonnes carbon dioxide equivalent, which represent about 10–13% of global emissions. Here, we review the role of the biogas sector in capturing methane and mitigating carbon emissions associated with biogas outputs. Since biogas impurities can cause severe practical difficulties in biogas storing and gas grid delivering systems, we present upgrading technologies that remove or consume the carbon dioxide in raw biogas, to achieve a minimum of 95% methane content. We discuss the role of hydrogen-assisted biological biogas upgrading in carbon sequestration by converting carbon dioxide to biomethane via utilising hydrogen generated primarily through other renewable energy sources such as water electrolysis and photovoltaic solar facilities or wind turbines. This conceptual shift of 'power to gas' allows storing and utilising the excess of energy generated in grids. By converting carbon dioxide produced during anaerobic digestion into additional biomethane, biogas has the potential to meet 53% of the demand for fossil natural gas. We also evaluate the role of digestate from biogas systems in producing biochar, which can be used directly as a biofertiliser or indirectly as a biomethanation enhancement, upgrading, and cleaning material.

Keywords Climate change · Net-zero · Biogas · Anaerobic digestion · Hydrogen economy · Ukraine conflict

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Introduction

The 2022 Ukraine conflict has put pressure on gas supplies, highlighting the need for more local energy resources. Renewable green energy transitions are necessary to mitigate global climate change and reduce carbon emissions worldwide (Levenda et al. 2021; Chen et al. 2022a). Bioenergy generation via anaerobic digestion of biomass in conjunction with carbon capture and storage or utilisation is one of the most frequently discussed negative emissions tools in recent years (Al-Wahaibi et al. 2020). Collecting all available wastes and anaerobically digesting them for biogas generation can reduce greenhouse gases emissions by 3.29 to 4.36 billion tonnes of carbon dioxide equivalent, which corresponds to 10–13% of the global greenhouse gas emissions from renewable bioenergy production, crop burning, evaded emissions management, landfill gas, deforestation, and fertiliser production emissions. Globally, the potential

for energy generation from currently available and recovered feedstocks ranges between 10,100 and 14,000 terra watt-hours. That energy could account for approximately 6–9% of primary energy consumed or approximately 23–32% of coal consumed globally. Anaerobic digestate as a soil biofertiliser has the potential to replace 5–7% of chemical fertiliser and fertilise over 80 million hectares of land (Jain et al. 2019; IEA 2018a).

Carbon dioxide sequestered or removed from raw biogas would reduce carbon dioxide emissions from anaerobic digestion systems, contribute to greenhouse gas mitigation, and have a more beneficial environmental effect. Thus, purification or upgrading of biogas is critical to removing carbon dioxide, increasing biogas electricity transmission efficiency, achieving optimal standardisation for direct integration into natural gas grids, and facilitating biogas transportation and storage, thereby optimising the function of renewable biogas as a fossil fuel substitute (Khan et al. 2021). In this regard, biological upgrading of biogas has been recognised as a simple and sustainable method for increasing the methane content of generated biogas while also stabilising wastes (Treu et al. 2018). The biological biogas upgrading technology is a hydrogen-assisted process that aims to sequester carbon dioxide by converting carbon dioxide to biomethane using hydrogen generated primarily from affordable renewable

energy sources such as water electrolysis, photovoltaic solar facilities, or wind turbines, thereby forming a new energy shifting technology called power to gas shift (Fu et al. 2021; Zhu et al. 2020a; Zabranska and Pokorna 2018).

In this review, we proposed that hydrogen transportation costs can be avoided if the applied hydrogen is derived from surplus hydrogen produced by water hydrolysis or wind or solar energy close to the biogas plant, which would provide sustainable, environmentally friendly, and cost-effective tools for upgrading biogas and sequestering carbon dioxide. Additionally, the digestates produced by the biogas system are massive, typically exceeding the required land area, and are stored in open tanks for an extended period, emitting greenhouse gases and polluting the environment. As a result, anaerobic digestion can be used in conjunction with biochar to establish circular multiple benefit concepts.

Therefore, several benefits can be obtained from biogas upgrading via biohydrogen injection and digestate-converted biochar recycling as shown in Fig. 1: (1) capturing and utilising upgraded biomethane ensures greenhouse gases zero-emission, thereby mitigating climate changes and global warming; (2), pyrolysis of digestate to biochar may ensure carbon sequestration, and hence, carbon zero-emission to the environment can be verified; (3), recycling biochar within the anaerobic reactor can improve biomethane upgrading
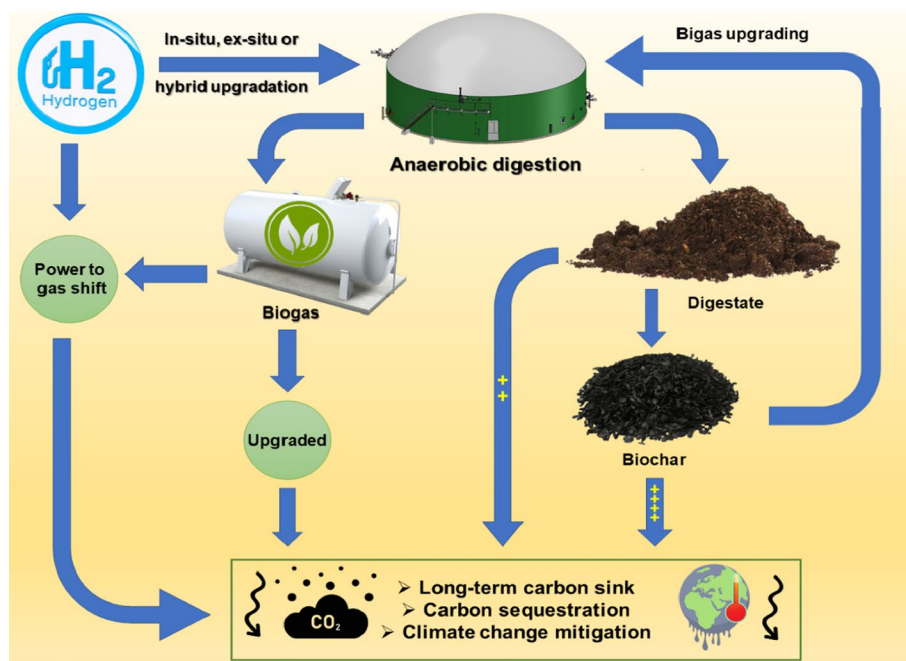
Fig. 1 System connecting anaerobic digestion, biochar, and dihydrogen to achieve carbon sequestration and climate change mitigation tool. Surplus hydrogen derived from water hydrolysis, wind, or solar electricity energy can be introduced into the anaerobic digestion system as a hydrogen-assisted biological biogas upgrading mean that can convert the carbon dioxide into biomethane to achieve new energy shifting technology called power to gas shift, offering sustain-able, environmental, and cost-effective tool to upgrade the biogas and sequester the carbon dioxide. In addition, the utilisation of digestate from the biogas system for biochar production would act as a long-term carbon sink that would be a useful tool for carbon sequestration and can be recycled inside the biogas system as a biomethanation enhancement and biogas impurities purification material, achieving circular approach

by boosting biomethane, enhancing microbial growth, and absorbing undesirable products (hydrogen sulphide and ammonia); (4), biochar produced from digestate can be easily stored and transported to the land for soil amendment purposes or making biochar-based fertiliser, along with other carbon sink applications; and (5) besides achieving the concept of "net-carbon zero emissions", economic and sustainable outcomes can be accomplished.

Biogas yield worldwide

Climate change directly impacts people's health, livelihoods, and well-being (Manning and Clayton 2018; Agha-Kouchak et al. 2020). In 2018, the world experienced over 300 climate-related natural disasters, affecting over 68 million people, resulting in approximately 131.7 billion dollars in economic losses, of which floods, storms, droughts, and wildfires accounted for 93%. The economic losses caused by wildfires in 2018 are nearly identical to the cumulative losses caused by wildfires over the last decade. Additionally, water, food, health, infrastructure, human habitat, and ecosystem are identified as the most vulnerable sectors to the climate crisis (Fawzy et al. 2020). As a result, climate change mitigation is critical to avoiding the worst of these consequences.

Greenhouse gas emissions, particularly carbon dioxide, are the primary cause of global climate change (Ritchie and Roser 2020). Global carbon dioxide emissions from fossil fuels were approximately 6 billion tonnes in 1950 and had exponentially increased to 34.81 billion tonnes in 2020 (Ritchie and Roser 2020), accounting for approximately 89% of the global carbon dioxide emission (Rogelj et al. 2018). As a result of global warming, 195 countries committed to the Paris Agreement in 2016 as a plan for global warming countermeasures. The agreement aims to keep the global temperature increase this century below 2℃ and to pursue efforts to limit global temperature to 1.5℃ by 2050. This would be accomplished by developing technologies with net-zero carbon emissions and reducing global greenhouse gas emissions (Osman et al. 2021a).

Renewable energy is the fastest-growing sector globally, with a projected 30% share of electricity in 2023, up from about 25% in 2018 (IEA 2018b). Additionally, by 2040, renewable energy is expected to account for two-thirds of global capacity for electricity production (IEA 2018b). By 2050, bioenergy sources such as biogas, biomethane, liquid biofuels, and solid biomass will account for 25% of the total primary energy supply (IRENA 2021). The share of bioenergy is equivalent to 460 megatonnes of oil in 2016 and is estimated to increase by 16.5% by 2023 (Xue et al. 2020); in addition, bioenergy share is estimated to be around

three-quarters of renewable energy worldwide, of which biomass represents more than the half (Xue et al. 2020).

Biomass has the potential to significantly increase energy supply in several countries with high energy demands, such as China and India (IRENA 2019). However, the utilisation rate for bioenergy production is different. For instance, from 1.4 gigatonnes of generated crop residues, about 900 million tonnes are annually utilised in China, as well as around 44% of the 2.05 gigatonnes of livestock and poultry manure are utilised (Xue et al. 2020). In comparison, Europe consumes approximately 1.2 gigatonnes of manure per year from a total of 1.35 gigatonnes generated (Scarlat et al. 2018a). Theoretically, in China, around 74 billion cubic metres of upgraded biogas (biomethane) can be produced from the 1.4 gigatonnes of wastes annually; however, the actual biogas produced in China is around 15.8 billion cubic metres in 2015 (Xue et al. 2020).

In comparison, European Union produced about 16.08 billion cubic metres of biogas from manure only (Scarlat et al. 2018b), which is greater than the entire yield of China despite China's higher quantity of manure produced (Xue et al. 2020). The differences in waste utilisation between Europe and China are attributed to the robustness of biomass storage and transportation systems (Xue et al. 2020); besides, the bioenergy consumption rates in the European Union are much higher than that in China (Wang et al. 2016). This difference indicates that most energy has not been utilised with contemporary biogas biotechnologies; however, China could potentially become a global leader in bioenergy consumption by implementing carbon sequestration and decarbonisation policies. As a result, standardisation of storage and transportation systems is critical to ensuring an adequate supply of biogas systems, as is optimising the use and development of biogas sectors.

Globally, there are 132,000 small to large scale biodigesters and around 50 million operating micro-scaled biodigesters that are primarily used for heating or cooking (Jain et al. 2019). The International Renewable Energy Agency's data demonstrate that the worldwide electricity produced from biogas was 46,243 gigawatt-hours in 2010 and nearly doubled to 91,819 gigawatt-hours in 2019 (IRENA 2019). This demonstrates a phenomenal growth trend in biogas production via anaerobic digestion. The anaerobic digestion system has the potential to generate 10,100–14,000 tera watt-hours of energy, which is equivalent to 6–9% of global primary energy consumption or approximately 32% of global coal consumption if the majority of wastes are utilised (IEA 2018a). Despite the growth in biogas production, approximately 407 tera watt-hours of biogas were produced globally in 2018, representing a small fraction of the biomethane energy potential estimated by the International Energy Agency, of which Europe, China, the United States

of America, and the rest of the world produced, respectively, 209, 84, 42, and 47 tera watt-hours (WBA 2021).

Europe is the world leader in biogas generation and utilisation, with 17,783 plants producing approximately 30 billion cubic metres per year (more than half of the world's total) and 63,504 gigawatt-hours biogas-derived electricity in 2018 (Lai et al. 2021). Around 74% of biogas generated in Europe comes from anaerobic digestion of agricultural residues, manure, and energy crops; 17% comes from landfills; and 9% comes from wastewater treatment plant sludge (Scarlat et al. 2018b). In 2015, European biogas was primarily used to generate 61 tera watt-hours of electricity and 127 terajoules of heat (50% biogas consumption). Additionally, Europe is the world's largest producer of biomethane used as a transportation fuel, with 697 biomethane filling stations supplying 0.16 billion cubic metres of biomethane to vehicles in 2015 as a transportation biofuel (Scarlat et al. 2018b).

On a regional scale, Germany, China, and the United States are the largest biogas-producing countries, with 328 picojoules, 272 picojoules and 156 picojoules, respectively (Lu and Gao 2021). Overall, Germany leads Europe's biogas sector with 10,971 large-scale plants, followed by 1665 biogas plants in Italy, 742 in France, 632 and 613 plants in Switzerland, and the United Kingdom, respectively (EBA 2019). Additionally, 110,448 biogas operations in China, of which 6972 large-scaled (2015), 2200 anaerobic biodigesters with 977 megawatts installed capacity in the United States, 180 digesters in Canada with 196 megawatts installed capacity, and 300 megawatts installed biogas capacity in India (Jain et al. 2019). The details of biogas plants and their production are listed in Table 1.

Biomethane conversion from biogas is a developed technology that aims to use biomethane as a vehicle fuel or to inject biomethane into regional gas grids. Alternatively, capturing biomethane and carbon dioxide during the establishment of biogas plants serves as a carbon sequestration tool. Certain plants begin to utilise the carbon dioxide captured in greenhouses, food processing plants, and beverage co-facilities. Of the 700 biogas upgrading plants worldwide, 570 are in Europe, with 203 in Germany, 96 in the United Kingdom, 69 in Sweden, 53 in the Netherlands, and 47 in France. Outside of Europe, there are approximately 50 upgrading plants in the United States, 25 in China, and 20 in Canada, as well as a few in Japan, India, Brazil, and South Korea (Jain et al. 2019; Bioenergy 2020).

According to the International Energy Agency Bioenergy Task 37-member countries, the biomethane markets in some countries like Germany, Sweden and the United Kingdom have extended, and the generated biomethane is used mainly for electricity and heat production, along with fuel for the vehicle. Countries have various financial support systems, such as tax exemptions, feed-in tariffs, and investment grants. A linear correlation between the way biogas is

utilised and the financial support system is obvious in the Task 37-member countries. In countries such as Germany, the United Kingdom, and Austria, the feed-in tariffs support for electricity generation has resulted in the use of almost all biogas in electricity generation, while the tax exemption system in Sweden encourages biomethane utilisation as a vehicle fuel. Other countries have developed financial support mechanisms for gas injection into their gas grids (Bioenergy 2022).

Energy transition and greenhouse gas mitigation potential from the anaerobic digestion process in wastewater treatment plants are critical. Global wastewater production is estimated to be 309.52 gigatonnes per year. Currently, the energy-consuming in the conventional activated sludge process is about 0.26–0.68 kilowatt-hour (kW-h)/cubic metre of the wastewater treatment process, with an average of 0.47 kW-h/cubic metre (Zhang and Ma 2020). If 40% of inflowing waste sludge's chemical oxygen demand is converted into methane via anaerobic digestion with a recoverable energy rate of 13.91 kilojoules (kJ)/g methane chemical oxygen demand, the recoverable energy unit via anaerobic digestion of waste sludge could be estimated as 500 g/cubic metre × 0.40 × 13.91 kJ/g = 2782 kilojoules/cubic metre. At an electricity conversion efficiency of 35%, total electrical energy of about 973.7 kilojoules/cubic metre (i.e. 0.27 kW-h/cubic metre) would be ultimately produced via anaerobic digestion of waste sludge meanwhile 1808.3 kJ/cubic metre would be harvested as heat from methane combustion. As discussed above, about 0.27 kW-h/cubic metre of electrical energy could be recovered through anaerobic digestion of waste sludge, indicating that the energy recovered in a current wastewater treatment plant could offset about 50–60% of total input energy (electricity only) consumption (Zhang and Ma 2020).

On the other hand, Pabi et al. (2013) reported that the wastewater treatment plants typically consumed about 4% of total electricity demand in the United States, accounting for more than 30% of total wastewater treatment costs. Nevertheless, the energy content of 13 MJ/kg chemical oxygen demand in wastewater sewage sludge (Gandiglio et al. 2017) is 3–5 times that of the total energy required for wastewater treatment. Consequently, effective utilisation of anaerobic digestion's biogas in wastewater treatment plants would facilitate a hypothesis transfer in wastewater treatment plants from an energy-intensive industry to an energy-positive or energy-neutral industry, thereby enhancing the economic development for the water treatment process.

Germany

In Germany, the overall renewable energy share would be increased to about 40–45%, 55–60%, and 80% by 2025 2035, and 2050, respectively. The biogas plants in Germany

Table 1 Biogas yields in some countries

Country	Biogas number					Total biogas production	Majority of biogas source	Biogas utilisation			
	Total	Wastewater treatment plant	Biowaste	Agriculture residues treating plants	Industrial treating plants and landfill			Electricity	Heat	Vehicle	Biomethane
Germany	10,551	1271	292	8400	280	54.1 terra watt-hours/year	Agriculture	31,706 gigawatt-hours/year (62.7%)	18,000 gigawatt-hours/year (35.6%)	884 gigawatt-hours/year (1.7%)	150,000 normal cubic metres/hour biomethane
United Kingdom	685	170	127	342	46	955 megawatts	Agriculture	702 megawatts	0.4 megawatts thermal	Not reported	96,171 normal cubic metre/hour
Sweden	282	134	36	54	58	2.16 terra watt-hours/year	Biowaste	40 gigawatt-hours/year (2%)	400 giga watt-hours/year (19%)	1401 gigawatt-hours/year (65%)	Not mentioned
Brazil	638	57	Not reported	503	78	11.7 terra watt-hours/year	Sanitary landfills and wastewater treatment plants (73%)	8500 gigawatt-hours/year (73%)	950 gigawatt-hours/year (8%)	Not reported	2140 (19%)
Norway	162	27	6	6	81	782 gigawatt-hours/year	Wastewater treatment plants	35 gigawatt-hours/year (5%)	168 gigawatt-hours/year (25%)	337 gigawatt-hours/year (51%)	Not mentioned
Switzerland	434	271	29	112	22	1.519 terra watt-hours/year	Wastewater treatment plants	372 gigawatt-hours/year (25%)	345 giga watt-hours/year (23%)	42 gigawatt-hours/year (3%)	Not mentioned
*France	687	88	16	390	193	3.933 terra watt-hours/year	Agriculture and landfill	1833 gigawatt-hours/year	1694 gigawatt-hours/year	Not reported	406 gigawatt-hours/year
*Denmark	172	51	Not reported	86	35	3.723 terra watt-hours/year	Agriculture	619 gigawatt-hours/year (17%)	1101 gigawatt-hours/year (29%)	Not mentioned	2003 gigawatt-hours/year upgraded (54%)
*Korea	119	36	55	7	21	2.815 terra watt-hours/year	Biowaste	1112 gigawatt-hours/year (39.5%)	667 gigawatt-hours/year (23.7%)	118 gigawatt-hours/year (4.2%)	601 gigawatt-hours/year (21.3%)

Overall, Germany leads the European biogas sector with 10,551 plants, followed by 687 in France, 685 in the United Kingdom, and 434 in Switzerland. Most biogas originated from agricultural sources, and the biogas generated is mainly utilised for electricity and heat. All countries according to Bioenergy (2022), while *is according to Bioenergy (2019)

mainly treat wastewater treatment plants (1271), biowaste (292), agriculture (8400), biomethane (232), and landfill (280). The 10,551 biogas plants in Germany are generating 33,600 and 20,500 GW-h/year) of electricity and heat, respectively. Based on the legalisation of using cultivated biomass for energy targets, the total calculated biogas potential for energy supply fluctuates between 155 and 265 TW-h/year. About 30% of the estimated potential is presently used for biogas production in Germany. In 2020, most biogas generated in Germany was applied for electricity (62.7%) and heat production (35.6%), though biomethane utilisation as an automobile fuel improved from 389 to 884 GW-h/year in 2019 and 2020, respectively. The number of filling stations delivering biomethane has risen from 100 in 2018 to approximately 550 stations in 202 (Bioenergy 2022).

The United Kingdom

The number of biogas plants in the United Kingdom is 170, 127, 342, and 46 for wastewater treatment plants, biowaste, agriculture, and industrial wastes, respectively. 120 biogas plants from the 685 anaerobic digestion plants in the United Kingdom inject their biomethane into gas grids (Bioenergy 2022). The overall biogas plants rapidly increased from 200 in 2010 to 650 in 2017, followed by very slow progress in the last years. There are 558 anaerobic digestion plants in the United Kingdom that generate electricity, 120 biogas plants for biomethane, and 7 plants only producing heat in 2020, in addition to 10 vehicle filling stations delivering biomethane (share of 67%) with compressed natural gas blended from the gas grid. The United Kingdom government implemented a 2050 net-zero carbon strategy by 2033–2037 (Bioenergy 2022). This challenging task to meet the United Kingdom's carbon net-zero target pushed the government and stakeholders toward decarbonising heat by applying anaerobic digestion-based biogas plants, ensuring carbon saving over natural gas.

The United Kingdom launched several initiative policies to address the decarbonisation of heat targets, such as the Sustainable Farming Incentive, Green Gas Support Scheme, and the Slurry Investment Scheme to further develop the biogas area (Bioenergy 2022). For instance, the green gas support scheme encourages injecting biomethane from anaerobic digestion into the gas grids. This support policy aimed to provide 2.8 TW-h of renewable heat/year by 2030. The green gas support scheme provides a 5.51 £/kW-h for the first 60 GW-h and 3.53 £/kW-h for the next 40 GW-h, then 1.56 £/kW-h for the rest of the biomethane production as a fixed tariff rate for biomethane uses for 15 years to increase the investment targets to this project. The new Green Gas Levy will provide £150 million per year to the anaerobic digestion industry, which would support 45 new biomethane plants (Bioenergy 2022).

In conclusion, the long-term carbon net-zero recognises anaerobic digestion-based biogas plants as a useful organic waste treatment alternative for the renewable production of heat fuel or electricity, and biogas' role in facilitating the decarbonisation of the gas grid is promising. In addition, the effluent nutrient-rich digestate has been recognised as a potential strategy to reduce greenhouse gas emissions (Bioenergy 2022). The biogas production in the United Kingdom is listed in Table 1.

France

The French Environment and Energy Management Agency aims to generate 70 terra watt-hours biogas annually by 2035. The vision is to establish 400 biogas plants every year. The generated energy from biogas plants represents 47% converted into electricity, 43% into heat and about 10% into biomethane. In 2017, 44 out of 47 upgrading plants injected biomethane into grids, generating 406 GW-h. In 2050, the vision is to produce 100 terra watt-hours/year (Bioenergy 2019).

Sweden

The Swedish climate and energy goals drive expanded renewable energy use, particularly in the transportation sector. The local 2020 renewable energy targets were achieved, with 50% overall energy utilisation and 10% transport goals. The Swedish Gas Association established a "Green gas 2050" vision that includes targets of 50 terra watt-hours renewable gas production by 2050, contributing to fossil-free land transportation, climate-neutral industry, fossil-free electricity and heat generation, and cleaner shipping (Bioenergy 2022).

The biogas plants in Sweden mainly treat wastewater treatment plants (134), biowaste (36), agriculture (54), industrial (7), and landfills (51). The 282 biogas plants in Sweden generate an overall 2.16 terra watt-hours of biogas in 2020. The generated biogas originated mostly from various co-digestion residues and bio-waste (52%), and sewage sludge (33%). Recently, 68 biomethane upgrading plants generating more than 1.3 TW-h biomethane per year, in addition to two biomethane liquefaction plants producing 78 gigawatt-hours/year, were installed (Bioenergy 2022).

Swedish biogas uses in transportation have increased promptly; however, biogas utilisation for heating has declined. Whereby 65% of the generated biogas was upgraded and used primarily as fuel for transportation due to a satisfactory financial support system. Biomethane use as vehicle gas steadily increased until 2014 and nearly stabilised at about 1500 gigawatt-hours. Biomethane utilisation accounted for over 95% of automobile gas usage in 2019–2020. Accordingly, the number of gas filling stations

has risen from below 20 in 2000 to around 200 filling stations in 2021, along with 60 private stations. During the same period, gas-dependent vehicles have increased from only a few hundred to about 53,982 in 2019, from which 2618 buses (representing 15% of all overall buses) and 1034 trucks, while the remaining number was for passenger and light cars (Bioenergy 2022).

Brazil

In Brazil, the biogas sector has increased in the last few years, with a total of 638 biogas plants in 2020 producing about 1.8 billion normal cubic metres/year (11.7 TW-h/year). The agricultural biogas plants were dominant (79%), yet almost of the biogas produced (73%) originated from sanitary landfills and wastewater treatment plants. The potential biogas generation from biomass has likely to be 82.58 billion normal cubic metres/year; hence 2% of the biogas potential is currently utilised (Bioenergy 2022). Most gas generated is used for electricity production (73%), while 8% for heat generation in 2020. The biomethane produced from the 8 biogas upgrading plants is utilised as a vehicle fuel and produces electricity. Brazil dedicated greenhouse gas emissions reduction by up to 43% by 2030, so biogas plants will contribute to meeting the carbon emissions reduction (Bioenergy 2022).

Denmark

Denmark's vision is to establish free fossil fuels dependency concept by 2050. Biogas production will assist in converting the fossil-dependent transport sector into a green-dependent (Bioenergy 2019). Biogas production has received considerable attention in the last few years as a significant and economical tool for mitigating climatic change and sequestering carbon dioxide emissions from the agricultural, transport, and energy sectors. Hence, the 172 biogas plants operating in Denmark in 2018 produced 3.7 TW-h energy. Almost of plants are agricultural (85%), either centralised biogas plants in farm clusters or on single farms. In 2018, 34 upgrading biogas plants were established to upgrade 54% of the generated biogas in Denmark. Besides, 29, 18.61, and 17% of the biogas produced were used for heat, biomethane, and power generation, respectively. The number of biomethane filling stations was 18 in 2018. Biomethane is anticipated to share by 30% in 2023 and 100% by 2035 as green utilised gas. Biomethane production would contribute to achieving the national climate vision and meeting the Paris Agreement commitment, as well as biomethane may be stored in present gas networks, offsetting other wind and green solar energies at no extra cost. The compressed upgraded biomethane utilisation for city trucks and busses is also growing to avoid

pollution and to meet economic incentives compared with imported diesel (Bioenergy 2019).

Republic of Korea

In Korea, 119 biogas plants are operating to produce 2815 GW-h per year of biogas. Biogas contributes from 31.8% (landfill), 22.4% (sewage sludge), and 45.5% (biowaste). The most important feedstock of biogas comprises food waste, food waste leachate, and other co-substrates. About 1112 GW-h (39.5%) of the biogas generated is utilised for electricity production, and 667 GW-h (23.7%) is used for heat production. Around 4.2% of the generated biogas is utilised for biomethane, while compressed gas supplies 39,081 buses as a fuel, with 201 gas filling stations reached, of which 6 biomethane filling stations supplying 0.2% of the total number of buses (Bioenergy 2019).

United States

Nearby over 2200 digesters produce biogas all over the United States, including 250 on-farm digesters, 1269 digesters treated water resources, 66 food treated digesters, and 652 landfill gas facilities. The potential biogas industry growth in the United States is huge, with possible 14,958 more sites ready for expansion nowadays, involving 8574 dairy, swine, and poultry and 3878 water treatment amenities, in addition to 2036 food scrap industries. The new biogas systems would generate 103 trillion kilowatt-hours of electricity/year and decrease the emissions, equal to the removal of 117 million customer vehicles from the street (ABC 2022).

According to the Environmental Protection Agency, manure-based anaerobic digestion systems increased from 169 in 2010 to 317 in 2021. The biogas plants include 112 cornered lagoons, 99 plug flow, 91 complete mix digesters, and 15 others. In 2021, the manure-based anaerobic digesters generated 1.59 million megawatt-hours of energy compared to 594.8 million kilowatt-hours equivalent in 2010 (EPA 2021). In addition, AgSTAR's *Market Opportunities for Biogas Recovery Systems* at United States *Livestock Facilities stated* that dairy and swine operations could produce 15.84 million megawatt-hours of electricity per year, corresponding to over 2009 megawatts of electrical grid capacity or around 5.4 million British thermal units of displaced fossil fuel use. The AgSTAR also estimated that the methane generation potential of swine and dairy farms is sufficient to heat more than 2.7 million homes or produce over 8 billion pounds of compressed natural gas annually (equal to 1.3 billion gallons of diesel), adequate to fuel approximately 150,000 trucks (EPA 2021).

Summary

Renewable energy share is nearly 19% of the worldwide energy mix, with an estimated increase of 50% by 2050. Bioenergy is shared by up to 10% of the worldwide primary energy supply with an expected increase to 60% owing to the potential impact of biogas production to diminish the fossil fuel uses and reduce greenhouse gas emissions by achieving carbon net-zero emissions. Europe is leading in biomethane production, so following Europe in creating sufficient biogas technologies to effectively utilise bioenergy is preferable. With increasing feedstock utilisation, the anaerobic digestion can produce over 14,000 terra watt-hours of energy, equivalent to 32% of the world's coal consumption by 2050. Increased use of bioenergy alongside other renewable sources will shift the world from coal-dependent to green energy-dependent. To achieve those goals, the government or decision-makers should encourage the biogas system establishment by launching several biogas support initiatives and other forms of tax exemption policies.

Role of biogas systems in climatic change mitigation

Role of biogas plants in greenhouse gases reduction

Anaerobic digestion of biomass is one of several waste-to-energy transmission technologies that can generate clean bioenergy in the form of biogas. This process can produce bioenergy from a variety of organic wastes, including agricultural residues, livestock manure, food waste, wastewater sludge, and macroalgae (Farghali et al. 2019; Yuhendra et al. 2021; Muhammad Nasir et al. 2012). Biogas is one of the most sustainable energy sources available for reducing reliance on and consumption of fossil fuels, reducing carbon emissions, alleviating the current fossil fuel crisis, avoiding deforestation, reducing harmful wastes, and enhancing soil fertility. Additionally, as previously discussed, biogas's diverse end-use capabilities and high utilisation efficiency are advantageous.

Carbon capturing and storage technologies can actively revert greenhouse gas emissions by locking the carbon contained in bio-carbon dioxide away from the atmosphere. Anaerobic digestion plants have the potential to become a carbon hub, concentrating atmospheric carbon within a digester and preparing carbon for permanent storage within materials or underground. The global industry would generate over 12,500 terra watt-hours of biogas at full capacity, equivalent to 1560 million tonnes of carbon dioxide (WBA 2021). Incorporating anaerobic digestion technologies into establishing a global green economy achieves both decarbonisation of waste management and defossilisation of fossil fuels, the matter that meets nine of the seventeen Sustainable Development Goals (SDGs) by 2030 (WBA 2021).

Collecting all available wastes and anaerobically digesting them for biogas production is expected to reduce greenhouse gas emissions by 3.29–4.36 billion tonnes of carbon dioxide equivalent, or 10–13% of global greenhouse gas emissions from renewable bioenergy production, crop burning, evaded emissions management, landfill gas, deforestation, and fertiliser production emissions, as seen in Table 2.

Biomass can be converted to a variety of renewable energy sources, including electricity, biogas, and liquid biofuels, thereby promoting environmental protection, public health, and sustainability (Osman et al. 2021b). For instance, the biogas produced by a single household-based biogas reactor can replace nearly 1.5 tonnes of straw and firewood combined each year. This could reduce two tonnes of carbon dioxide emissions per year (Xue et al. 2020). Additionally, capturing carbon from various biomass through anaerobic digestion has the potential to reduce 3–4 gigatonnes of carbon dioxide, or more than 10% of global greenhouse gas emissions (Jain et al. 2019). Compressed biogas reduced greenhouse gas emissions by 93–131% to fossil diesel (van den Oever et al. 2021).

The reduction in methane emissions from a biogas system (anaerobic lagoon) was estimated compared to a manure storage tank or pond on a 500-cow dairy farm in California. The baseline manure management process emits 82.6 tonnes of methane per year, which is equivalent to 2064 tonnes of carbon dioxide equivalent per year; however, biogas capturing reduces methane emissions by 179.4 tonnes per year (equivalent to 4485 tonnes of carbon dioxide equivalent per year). Additionally, utilising biogas for electricity generation could reduce 828 tonnes of carbon dioxide equivalent per year from conventional power generation sources, as fewer fossil fuels are combusted by electric power plants (EBA 2018).

In 2020, the United States 317 manure-based anaerobic digesters reduced greenhouse gas emissions by 5.29 million metric tonnes of carbon dioxide equivalent ($MMTCO_2$eq), resulting from 4.44 $MMTCO_2$eq direct methane reductions + 0.83 $MMTCO_2$eq emissions avoided. At the same time, an increased reduction from direct (5.07) and indirect emissions (0.88) of 5.95 $MMTCO_2$eq was achieved from livestock farms-based anaerobic digesters in 2021. Besides, between 2000 and 2020, livestock manure-based digesters established in the United States have reduced direct and indirect emissions by 48.3 $MMTCO_2$eq (EPA 2021). As a result, biogas generated through anaerobic digestion significantly impacts energy resources and carbon emissions to the environment.

Olesen et al. (2021) analysed and quantified the potential of the biogas system to abate greenhouse gas emissions. The authors selected five biogas production models with varying

Table 2 Various biomass potentials for bioenergy production through the anaerobic digestion process (Jain et al. 2019; IEA 2018a)

Biomass	Global production	Global energy generation potential	Coverage electricity demand	Biomethane upgrading substitutes natural gas	Greenhouse gases mitigation potential	Digestate potential
Livestock manure (current situation)	1.7 billion cattle and buffalo (16 tonnes manure/head/year), 1 billion pigs (1.6 tonnes manure/head/year), 22 billion chickens (0.112 tonnes droppings/bird/year)	2600 to 3800 terra watt-hours	330 to 490 million people	250 to 370 billion cubic metres of biomethane	Reduce global greenhouse gases emissions by 930 to 1260 million tonnes of carbon dioxide equivalent/ year (about 13–18% of the existing livestock emissions)	10 billion tonnes of nutrient-rich digestate fertilisers
2050 situation	Growth rates are assumed to be 1.5% for poultry, 0.6% for cattle and buffaloes, and 0.5% for pigs	4798 terra watt-hours	Not available	Not available	1193 million tonnes of carbon dioxide equivalent emissions	Not available
Sewage (current situation)	7.5 billion in 2017, each one generates around 128 g of faeces/day	210 to 300 terra watt-hours	27 to 38 million people	22 to 32 billion cubic metres of biomethane	Can mitigate 75 to 100 million tonnes of carbon dioxide equivalent/annum	0.23 billion tonnes of digestate (substituting around 0.4–3% of global chemical fertiliser used)
2050 situation	Not available	385 terra watt-hours/year	Not available	Not available	95 million tonnes of carbon dioxide equivalent/year	Not available
Food wastes (current situation)	1.6 billion tonnes/year	880 to 1100 terra watt-hours	112 to 135 million people	Not available	510 to 560 million tonnes of carbon dioxide equivalent	Can replace 2–5% of the current worldwide synthetic fertiliser consumption
2050 situation	Not available	340 terra watt-hours/year	Not available	Not available	271 million tonnes of carbon dioxide equivalent/year	Not available
Crop residue wastes (current situation)	Varies depending on the type of crop (Harvesting and processing residues)	3080 to 3920 terra watt-hours	393 to 500 million people	300 to 380 billion cubic metres of biomethane per year	865 to 1100 million tonnes of carbon dioxide equivalent per year	Not available
2050 situation	Not available	5432 terra watt-hours/year	Not available	Not available	1063 million tonnes of carbon dioxide equivalent/ year	Not available
Energy crops (current situation)	Grow on 7% of agricultural land	3350 to 5000 terra watt-hours	Not available	330 to 490 billion cubic metres of biomethane	910 to 1350 million tonnes of carbon dioxide equivalent per year	Not available
2050 situation	Not available	4967 terra watt-hours/year	Not available	Not available	945 million tonnes of carbon dioxide equivalent/year	Not available

The key finding of the potential of anaerobic digestion currently and in the year 2050 as a target to decrease greenhouse gases releases, meet energy demand, natural gas usage substitute, and recover the digestate nutrient potential are listed. The current greenhouse gas emissions would be reduced by 3.29 to 4.360 gigatonnes carbon dioxide equivalent (representing 10–13% of the global greenhouse gas emissions). Energy production potential from the currently available and recovered feedstocks worldwide is 10,100–14,000 terra watt-hours. This energy can cover around 6–9% of the primary energy utilisation or 23–32% of the coal utilisation over the world. When utilised for electricity generation, energy would meet 16–22% of the world's electricity consumed. If the energy is used as biomethane, energy may replace 993 to 1380 billion cubic metres of natural gas, equal to 26–37% of the world's natural gas used. Applying anaerobic digestate in soil may substitute 5–7% of chemical fertiliser and can fertilise more than 80 million hectares of land

bioresources compositions to illustrate how Danish biogas technology is best applied. Additionally, they used various digestion "retention" times ranging from 45 to 90 days and assumed that the biogas was generated by centralised large biogas systems that converted biogas to biomethane with the same value as natural gas. The authors assumed that 50% of digested effluent is stored in solid-covered tanks for a period of 20 days prior to being transported to farms. The greenhouse gas and environmental effects were calculated using the greenhouse gas warming potentials of nitrous oxide and methane, which are 298 and 25 carbon dioxide equivalents, respectively.

Table 3 illustrates the greenhouse gas emissions associated with various assumed scenarios. The greenhouse gas emissions calculated include biogas energy production that offsets fossil fuel-based carbon dioxide emissions; 1% methane escape from biogas upgrading; methane emitted during biomass storage; and nitrous oxide emitted during storage, nitrate leaching ammonia volatilisation, and field application. Their findings showed a total greenhouse gas reduction of 65–106 kg of carbon dioxide equivalent per tonne of biomass at 45 days retention time in the modelled biogas systems. The major greenhouse gas offset was achieved by utilising biogas via a natural gas grid that replaces fossil fuel and methane-producing from conventional storage of wet biomasses.

In the United Kingdom, 67% rated greenhouse gas emissions as fairly important to extremely important when making decisions about their livestock, crops, and land in 2021. 56% of them have a strategy to mitigate their farm's greenhouse gas emissions. The most frequently cited actions for reducing greenhouse gas emissions on farms were waste recycling (83%), energy efficiency improvement (79%), and increased accuracy or efficiency in nitrogen fertiliser application (62%) (DEFRA 2021).

Role of biomethane as fuel in greenhouse gas reduction

Climate change can lead to massive irreversible and negative impacts on humanity and ecosystems, combined with immense socioeconomic consequences. Therefore, reducing anthropogenic greenhouse gas emissions resulting from the extraction and combustion of fossil fuels is critical (Gaulin and Le Billon 2020) to keep global warming lower than 1.5 °C, as indicated in the Paris Agreement. Transportation

Table 3 Greenhouse gas emitted from different biogas model plants (Olesen et al. 2021)

Biogas model	Deep litter and slurry	Straw and slurry	Deep litter, slurry, and maize ensilage	Organic waste, deep litter, and slurry	Organic waste, slurry, organic grass-clover, and deep litter
Total greenhouse gas reduction (kilogramme carbon dioxide equivalent/tonne biomass) at 45 days retention time	66.76	105.53	67.74	65.32	99.46
Total greenhouse gas reduction per gross energy produced (kilogramme carbon dioxide equivalent/gigajoule gross energy) at 45 days retention time	77.46	52.89	68.35	52.74	54.65
Total greenhouse gas reduction (kilogramme carbon dioxide equivalent/tonne biomass) at 65 days retention time	68.76	110.72	69.60	67.55	102.59
Total greenhouse gas reduction per gross energy produced (kilogramme carbon dioxide equivalent/gigajoule gross energy) at 65 days retention time	76.47	53.61	68.19	53.29	55.21
Total greenhouse gas reduction (kilogramme carbon dioxide equivalent/tonne biomass) at 90 days retention time	70.29	112.70	70.64	68.25	102.53
Total greenhouse gas reduction per gross energy produced (kilogramme carbon dioxide equivalent/gigajoule gross energy) at 90 days retention time	76.10	53.53	67.85	53.04	55.08

The total greenhouse gas emissions reduction of 65–106 kg carbon dioxide equivalent per tonne biomass at 45 days retention time in the modelled biogas systems was achieved. The slurry is a mixture of pig and cattle slurry; deep litter storage took place in covered heaps for five months; the energy crop is silage maize; and the organic waste included slaughterhouse waste, glycine, and biowaste from a household

of people and goods consumes enormous amounts of energy and is classified as an air polluter and a greenhouse gas emitter (Tian et al. 2018). Transportation accounts for 29% of global carbon dioxide emissions, the majority of which result from fossil-fuel combustion (EPA 2019).

Calculations of greenhouse gas emissions from the transportation of gas can be classified as tank-to-wheel, well-to-tank, the cascade steps required to generate fuel from primary source to delivery and vehicle refuelling, or well-to-wheel, which encompasses the entire fuel production life cycle from a primary source to end-use, as shown in Fig. 2. We can quantify the climate impacts of biogas systems at various stages of their life cycle by implementing the well-to-wheel concept. For instance, by using animal manure as a feedstock for the biogas system, avoiding the climate effects associated with traditional manure management is possible, in which methane is produced and emitted in an uncontrolled manner as a result of manure open storage. Alternatively, feeding manure into a biogas system that captures the methane produced and the carbon dioxide produced by the engine's methane combustion produces less greenhouse gas (Ammenberg et al. 2021). Thus, proper handling and treatment of manure, combined with proper fertilisation practices for the digestate, can result in a carbon–neutral lifecycle approach from a climate standpoint (Börjesson and Iverfeldt 2016).

In general, biomethane produced from wastes performs better in terms of climate and environmental performance than biomethane produced from energy crops, owing to the larger environmental problem associated with wastes. Typically, digestate benefits are not considered in life cycle assessments of biogas potentials that use biomethane for transportation, despite the significant indirect definite climate impacts associated with digestate produced by biogas sectors. Wherever digestates are applied to soils, they can be used in place of inorganic fertilisers (McCabe et al. 2020) that require vast quantities of natural gas.

Natural gas emits fewer greenhouse gases than diesel (Börjesson and Iverfeldt 2016; Speirs et al. 2020). However, this reduction is limited by methane leakage, the lower efficiency of gas engines compared to diesel engines, and the fact that natural gas is a fossil fuel. Additionally, the production of liquified natural gas resulted in greenhouse gas emissions (Stettler et al. 2019; Cooper et al. 2019). Technically, fossil-derived natural gas can simply be replaced with biomethane derived from renewable sources, either liquefied or compressed. The utilisation of biomethane in the transportation sector has several advantages: (1) biomethane is derived from organic biomass that has nearly identical properties to natural gas (Speirs et al. 2018); (2) biomethane from biogas plants can be produced and used locally, thereby avoiding the difficulties associated with natural gas importation, particularly in Europe (Eurostat 2022; Kampman et al. 2017); and (3) utilising biomethane in transportation systems would be a cost-effective way to reduce carbon emissions and combat climate change. Currently, the use of biomethane in vehicles is extremely limited, with Sweden leading the way in Europe.

The environmental merits of using biomethane as a vehicle fuel can be summarised as follow:

Prussi et al. (2020) found that biomethane derived from manure, whether liquefied or compressed, had the lowest well-to-wheel greenhouse gas emissions of 250 energy carriers for road transport studied, with renewable biomethane offering a net emission of -239.3 g carbon dioxide equivalent/megajoule fuel when the manure managed in a closed storage system. Similarly, Pääkkönen et al. (2019) concluded that by 2030, biomethane fuel shifting could support 50% of Finland's heavy-duty transportation sector. Additionally, the authors estimated a biomethane production cost of 81–190 €/
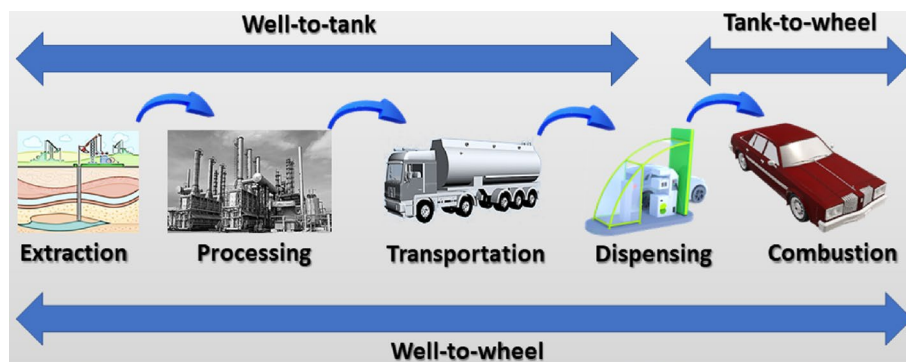
Fig. 2 Well-to-wheel concept. The greenhouse gas emissions calculations from the transportation sector of the gas sector can classify into tank-to-wheel, well-to-tank (the cascade steps to generate fuel from the primary source to fuel delivery and vehicle refuelling), well-to-wheel, which involve the whole fuel production life cycle to the end-user. Utilising biomethane for transportation can significantly reduce the consumption of fossil fuels, thereby lowering greenhouse gas emissions. Currently, engines in trucks and buses are adapted to utilise methane in order to meet environmental and climate goals

MW-h, which is comparable to diesel's consumer price of 152 €/MW-h. If all biomethane is used in heavy-duty transport, carbon dioxide emissions are reduced by 50%, reducing environmental impact and mitigating global warming (Lyng and Brekke 2019). Biomethane produced from manure has the potential to reduce greenhouse gas emissions by 1.0–2.6 kilogrammes carbon dioxide equivalent, resulting in a net negative emission across the well-to-wheel cycle of − 2.2 to − 0.6 kg carbon dioxide equivalent/km bus transportation. Additionally, replacing mineral fertilisers with biomethane derived from food waste would result in negative emissions of − 0.90 (Lyng and Brekke 2019).

The most frequently used feedstock for biomethane production is livestock manure, which accounts for 43% of the European transport sector (Baldino et al. 2018; Hijazi et al. 2016). Renewable biomethane use in road transports has the potential to reduce the greenhouse gas emissions by − 264 g carbon dioxide equivalent/megajoule (− 9.45 kg carbon dioxide equivalent/cubic metre) from the anaerobic digestion of livestock manure compared to emissions of 26, 68.6, 46.9, and 72 g carbon dioxide equivalent/megajoule from renewable solar power-to-gas (electrolysis and methanation) in 2030–2050, EU-28 Power production, grid average in 2030–2050, and Fossil fuel-derived fossil gas, respectively (Baldino et al. 2018). The lower rate of biomethane use in transportation compared to electricity and heat is due to the financial incentives associated with low carbon production approaches (Baldino et al. 2018). Ammenberg et al. (2021) and Long and Murphy (2019) reported that the utilisation of biomethane from grass silage and slurry met sustainability criteria for heat and transportation, emitting 27 and 22.95 g of carbon dioxide equivalent/megajoule biomethane, respectively, compared to 80- and 94-g carbon dioxide equivalent/megajoule for the fossil fuel comparator, implying a 66–76% reduction in emissions.

Cenex (2019) evaluated the performance of 20 biomethane-fueled gas vehicles in the United Kingdom. The findings indicated an 80% significant reduction in greenhouse gas emissions compared to diesel across all vehicle types; specifically, using fossil-derived natural gas would result in emissions savings of 13% to a 4% increase compared to diesel. If the trial vehicles travelled more than 2.2 million kilometres, they would save over 1400 tonnes of carbon dioxide equivalent from well-to-wheel. This is equivalent to the carbon dioxide emitted by 3150 trees over their lifetime, occupying an area roughly the size of 18 football pitches Ammenberg et al. (2021) concluded that using renewable biomethane as a vehicle fuel reduced the greenhouse gas by 80–90% according to different calculations compared to gasoline- and diesel-fueled vehicles, whereas fossil natural gas could reduce greenhouse gas emissions by 10–15% when compared to gasoline.

Atkins et al. (2021) demonstrated that using biomethane as a vehicle fuel could be a quick way to decarbonise heavy-duty trucks. Where a lifecycle assessment indicated that liquefied biomethane could reduce greenhouse gas emissions by 75% compared to a diesel standard, battery-electric and fossil natural gas could reduce greenhouse gas emissions by 49 and 8%, respectively. Due to the substitution of fossil fuels and avoided methane emissions, upcycling biomethane from sewer systems and waste grass anaerobic digestion resulted in net negative carbon dioxide equivalent emissions of 246%.

However, technical and energy barriers exist, such as the need to increase gas grid pressure to 240 bar for gas vehicle fueling, as well as the inefficiency of regional delivery, distribution, and charging processes for electric vehicles. Piechota and Igliński (2021) reported that compressed natural gas/biomethane is subject to more restrictions than liquefied petroleum gas. Where approximately 200 bar pressure is required to compress the volatile gas into costly tanks; in addition, biomethane fueled vehicles can travel approximately 400 kms on a single tank. The authors proposed liquefied biomethane as an optimised solution not only for the aforementioned technical barriers but also as a green energy generation, waste disposal, and negative carbon emissions technology with a 75–200% reduction in carbon emissions compared to diesel.

Numerous authors emphasised the environmental benefits of liquified biomethane over diesel in heavy-duty trucks (Shanmugam et al. 2018; Hagos and Ahlgren 2018; Gustafsson et al. 2021). Gustafsson et al. (2021) found a positive correlation between climate change mitigation and the proportion of biomethane in compressed natural gas versus fossil-derived natural gas or fossil diesel. In this context, a 50% biomethane content in compressed natural gas would reduce the climate change impact of vehicles by 35 and 45%, respectively, compared to fossil compressed natural gas and fossil diesel. A 100% biomethane share would mitigate climate change by over 65 and 75%, respectively, compared to fossil compressed natural gas and fossil diesel. If the anaerobic digestate is included, the climate change impact is reduced by 45–60% at a 50% biomethane content in compressed natural gas and by 95% at a 100% biomethane content (Gustafsson et al. 2021). Compared well-to-wheel scenarios to assess the environmental impacts of liquefied biomethane and liquefied natural gas over their respective life cycles. The authors concluded that liquefied natural gas could contribute 10% more to climate change than diesel. However, liquefied biomethane can significantly reduce environmental impact by 45–70% and 50–75%, respectively, compared to a diesel with manure-derived liquefied biomethane and food waste-derived liquefied biomethane. Additionally, when biogas digestate is used in place of chemical fertiliser, the climate benefits of using liquefied biomethane

as a diesel substitute are greater. Where liquefied biomethane derived from manure has the potential to reduce climate change by 100–125%, compared to 80–105% for liquefied biomethane derived from food waste (Gustafsson and Svensson 2021).

Role of biogas plants in emissions savings by displacing synthetic fertiliser with digestate

Fertilisers are primarily used on crops in order to meet global food demand. Intensive use of inorganic fertilisers has resulted in risks to human health and the environment, including a high carbon footprint, a high cost, nitrate leaching pollution, eutrophication risk in water bodies, decreased soil microbial activity, and soil organic carbon loss where inorganic fertilisers do not deliver the organic matter to the soil. From a soil health perspective, the application of inorganic fertilisers may reduce soil productivity, water retention capacity, salinity, alter soil minerals, and expose crops to various infections (Rahman and Zhang 2018).

The manufactured nitrogen cycle is unsustainable, with 30–40% of applied nitrogen being used by crops and thus entering the food chain, while the remainder is frequently lost to waterways or the atmosphere. Additionally, approximately 12.5% of the food chain's nitrogen is excreted by humans and returned to the atmosphere via aeration. This cycle typically consumes two litres of fossil fuel to generate one kilogramme of nitrogen, which is unsustainable (WBA 2021).

Mineral fertilisers are produced artificially by converting chemicals into usable forms. The Haber Bosch process is by far the most common, as the Haber Bosch process combines nitrogen gas from the atmosphere with hydrogen from fossil natural gas (methane) to form liquid ammonium. This process consumes a large amount of natural gas and energy. According to multiple studies, nitrogen fixation consumes approximately 3–5% of global natural gas generation and accounts for 1–2% of the global annual energy supply to produce approximately 450 million tonnes of nitrogen fertiliser (WBA 2021; Akbar et al. 2021).

Fertiliser consumption varies significantly across the globe. Brazil consumes approximately 186 kilogrammes per hectare of arable land, 503 kilogrammes per hectare for China, 165 kilogrammes per hectare for India, 231 kilogrammes per hectare of arable land for Indonesia, more than 1240 kilogrammes per hectare of arable land for Ireland, 71 kilogrammes per hectare of arable land for Morocco, 59 kilogrammes per hectare of arable land for South Africa, 138 kilogrammes per hectare of arable land for the United States, and 253 kilogrammes per hectare of arable land for the United Kingdom (WBA 2021).

Apart from the environmental and soil concerns raised by the use of fossil fuels to synthesise chemical fertilisers, the concern about the depletion of chemical fertilisers' global natural resources has increased; for example, phosphorus is expected to be depleted within 50 to 100 years, posing a threat to crop development and food security (Akbar et al. 2021). Within 30 to 40 years, the existing phosphorus supplies possibly would be insufficient to meet global agricultural demand (WBA 2021). Thus, sustainable and novel alternatives to the traditional use of chemical fertilisers are required.

To avoid the limitations mentioned above, the digestate output of anaerobic digestion can be used as a bio-fertiliser in place of inorganic fertilisers. Anaerobic digestate contains abundant nutrients, has excellent fertiliser potential, and has a significant global capacity (Kumar et al. 2020a).

Digestate is the term used to describe anaerobic digestion feedstock that has been degassed. Typically, scientists combine the suffix 'ate' to denote the result of an action; thus, the effluent of anaerobic digestion is digest–ate, the result of digestion. For every tonne of feedstock degassed in a digester, approximately 50–85% by weight of the feedstock is recovered as digestate. Anaerobic digestion treats nutrients that would have been disposed of in water bodies or landfills without anaerobic digestion. Digestate contains all of the nutrients contained in organic wastes. As such, digestate is known by various other names throughout the world, including biofertiliser and organic/natural fertiliser (WBA 2021). In general, digestate takes on various forms: With less than 5% total solids, the first form is comparable to livestock slurry. This material is extracted directly from the digester and may then be separated into two parts: A liquid biofertiliser in which the majority of the total solids have been separated and a solid biofertiliser in which the remaining dry matter has been retained.

Following solid–liquid segregation, the liquid fraction contains a higher nitrogen content (70–80% of total ammonium nitrogen) and a lower phosphorus content (35–45%), whereas the solid fraction contains a higher phosphorus content (55–65% of total phosphorus) and a residual of total ammonium nitrogen (20–30%) (Logan and Visvanathan 2019). Thus, the solid digestate fraction has a greater potential for soil improvement, whereas the fluid digestate fraction has a higher fertiliser potency.

Digestate has the potential to significantly reduce the demand for mineral fertilisers by recovering nutrients from organic wastes. As a result, digestate reduces our reliance on fossil natural gas, the need for the global transportation of these fertilisers, and all associated emissions. When one tonne of mineral fertiliser is replaced with a tonne of organic fertiliser, one tonne of oil, 108 tonnes of water, and 5–9 tonnes of carbon dioxide equivalent emissions are saved (WBA 2021). Likewise, Akbar et al. (2021) reported that digestate biofertilisers could substitute synthetic fertilisers in a natural, cost-effective, degradable, and

environment-friendly way; additionally, they promote mineral recirculation and increase global food safety potential by increasing crop productivity and soil fertility in a shorter period of time. Along these, anaerobic digestates provide several benefits, including a low carbon footprint, the ability to manufacture locally with relatively low manufacturing costs, and the ability to reduce greenhouse gas emissions. The United Kingdom frequently spends more than £250 million per year on synthetic fertilisers. The authors also noted that substituting digestate for one tonne of synthetic fertiliser saves nearly 108 tonnes of water and 4 tonnes of carbon dioxide equivalent.

Timonen et al. (2019) stated that considering the climate potential and credits of the anaerobic digestate is extremely important, the matter which is allocated mainly for energy production and use. The authors reported that the storage, transportation, and use of digestate biofertiliser on-farm produced more greenhouse gases than mineral fertiliser used on-farm. Nonetheless, total anaerobic digestion cycle emissions up to digestate utilisation were significantly reduced compared to mineral fertiliser generation and field application. By substituting digestate for inorganic fertiliser, the agroecosystem shifted from 2.7 tonnes of carbon dioxide equivalent/square metric hectare/year to 8.8 tonnes of carbon dioxide equivalent/square metric hectare/year carbon sink (Liu et al. 2015).

Challenges facing biogas systems

Numerous countries recognise the critical role of biogas system expansion due to the sustainability transition, energy demand, and climate change mitigation. Nonetheless, global biogas production is still relatively low, with only 5% of generated biogas being used to generate biomethane, whereas complete utilisation of organic wastes for biomethane generation could meet 20% of global gas demand (Golmakani et al. 2022; IEA 2020). Indeed, this sector faces several major obstacles limiting the biogas sector's ability to achieve the objectives mentioned above. The following are the major barriers to the widespread use of biogas globally: technical considerations, economic constraints, market opportunity, institutional constraints, and sociocultural considerations (Nevzorova and Kutcherov 2019). This section focuses on the technical barrier facing the widespread adoption of biogas plants.

Impurities in biogas are a significant issue, affecting both uptake and utilisation, as well as the performance of biogas equipment. Carbon monoxide, water vapour, hydrogen sulphide, nitrogen gas, oxygen, ammonia, and siloxanes, in particular, may present difficulties, including toxicity, corrosion, and a decrease in the heating value of biogas (Khan et al. 2021; Bragança et al. 2020).Hydrogen sulphide is frequently the cause of the corrosion, and hydrogen sulphide presence

in biogas at concentrations greater than 800 mg/l can result in unconsciousness, respiratory palsy, or even death. The siloxanes are converted to silica during biogas ignition, which initiates abrasion of the vehicle's spark plugs, cylinder heads, and valve heads. Water vapour in biogas causes severe corrosion when water reacts with hydrogen sulphide, ammonia, and carbon dioxide, resulting in the formation of acid (Golmakani et al. 2022; Muñoz et al. 2015). Besides, the presence of carbon dioxide in the biogas produced is another major challenge. Huang et al. (2016) reported that injecting raw biogas containing impurities into the gas pipe system decreased biogas heating value due to the presence of the carbon dioxide, could block the pipes network due to the existence of dust, and might damage and clog equipment and pipe systems due to the presence of liquid water, which may freeze at low temperatures. The presence of solid sol could block the compressors, valves, and other facilities. The hazards of impurities to biogas facilities summarise in Table 4.

Carbon dioxide is more abundant in biogas than any other impurity. The heating value of raw biogas is approximately 5000 kcal/normal cubic metre, with a methane content of 60% (volume/volume). After carbon dioxide and other impurities such as water and hydrogen sulphide are removed, the residual gas is identified as biomethane, which has properties similar to purified natural gas. As a result, this value would be increased to levels comparable to natural gas (8000 kcal/normal cubic metre) when biogas is upgraded to biomethane with a concentration greater than 96% (Tabatabaei et al. 2020). Hence, the calorific value and valorising degree of biogas are mainly dependent on the methane content; hence, removing carbon dioxide considerably improves the calorific value of biogas.

Among several technical difficulties, infrastructural difficulties are a frequent critical barrier. The limited transportation infrastructure for gas-fuelled vehicles, in particular, makes the transportation sector more difficult to reach a fossil-free vehicle station, restricting biogas expansion (Nevzorova and Kutcherov 2019). Additionally, the shortages of pipelines connected to the grid systems, as well as challenges such as suitable waste collection, waste segregation, and storage, impede the utilisation and distribution of biogas (Chien Bong et al. 2017; Mittal et al. 2018).

In Latin America, household biodigesters are completely reliant on locally generated organic waste. Nonetheless, feedstock collection, construction resources, and digestate flow are not always feasible in remote and rural areas due to difficult or lengthy transportation routes (Einarsson and Persson 2017; Garfí et al. 2016). Thus, establishing suitable land for installing biogas equipment alongside an adequate number of livestock to supply feedstock in rural and developing countries is critical. Other obstacles, such as a lack of available local biogas facilities, could increase imported equipment's investment and maintenance costs (Kamp and

Table 4 Impact of biogas contaminations on biogas utilisation (Golmakani et al. 2022; Muñoz et al. 2015; Franco-Morgado et al. 2018; Angelidaki et al. 2019)

Impurity	Potential impact	Biogas standards for grids or transportation use	Raw biogas
Hydrogen sulphide	Corrosion of compressors Engines and other gas storage containers damage (> 5 cubic centimetres/cubic metre) Formation of sulphur trioxide and sulphur dioxide during incineration triggering rusting, especially in the existence of water	< 5 mg/cubic metre	0–10,000 parts per million
Water vapour	Corrosion of compressors Engines and other gas storage containers damage due to acid formation when reacting with ammonia, carbon dioxide, and hydrogen sulphide	< 32 mg/normal cubic metre	5–10%
Carbon dioxide	Lowering the calorific value of the biogas Adversely influencing the engine's anti-knock features Corrosion action	< 2%	30–50%
Siloxanes	Form silicon dioxides and microcrystalline silica during incineration	5–10 mg/cubic metre	0–41 mg/cubic metre
Ammonia	Forms rust and corrosion after dissolving in water Emission problems Adversely influencing the engine's anti-knock features	< 3–20 mg/cubic metre	0–100 parts per million
Nitrogen	Reduces the calorific value of the biogas Adversely influencing the engine's anti-knock features Corrosive action	Not mentioned	0–3%
Oxygen	Corrosive Fooling in gas storage Explosive	< 0.2–0.5%	0–1%
Hydrocarbons	Cause engine's rust	Not mentioned	0–200 mg/cubic metre
Total chlorine as chloride ions	Cause engine corrosion	< 1 mg chloride ion/cubic metre	Not mentioned

Impurities in biogas, such as carbon dioxide, water vapour, hydrogen sulphide, nitrogen gas, oxygen, ammonia, and siloxanes, may present difficulties, including toxicity, corrosion, and a decrease in the heating value of generated biogas. Corrosion is frequently triggered by hydrogen sulphide. During incineration, the siloxanes are converted to silicon dioxides and microcrystalline silica. Water vapour in biogas causes severe corrosion when water reacts with hydrogen sulphide, ammonia, and carbon dioxide, resulting in acid formation. Additionally, the presence of carbon dioxide in biogas reduces biogas calorific value and has a detrimental effect on the engine's anti-knock characteristics

Forn 2016; Hoo et al. 2018). Additionally, a lack of knowledge about the proper use and fertilisation of digestate hinders the successful use of biofertiliser and biogas; thus, farmers should be trained and educated on the proper use of feedstock for biofertiliser production (Uddin et al. 2016). Continuous training in the operation and maintenance of digesters is critical to ensuring an efficient supply of energy to households, as is knowledge of the safety and limitations of biogas plants (Garfí et al. 2016). The detailed challenges associated with digestate utilisation are discussed in Sect. 2.

Summary

Carbon capture and storage technologies can actively revert greenhouse gas emissions by locking the carbon contained in bio-carbon dioxide away from the atmosphere. Anaerobic digestion plants have the potential to become a carbon hub, concentrating atmospheric carbon within a digester and preparing carbon for permanent storage within materials or underground. At full capacity, the global industry would generate over 12,500 terra watt-hours of biogas, equivalent to 1560 million tonnes of carbon dioxide. Additionally, by collecting all available wastes and digesting them anaerobically for biogas generation, greenhouse gas emissions can be reduced by 3.29 to 4.36 billion tonnes of carbon dioxide equivalent, or 10–13% of global greenhouse gas emissions. Due to the potential role of anaerobic digestion technologies in establishing a global green economy, both waste management decarbonisation and fossil fuel defossilisation would be achieved by 2030, enabling the implementation of nine of the seventeen Sustainable Development Goals (SDGs).

Transportation accounts for 29% of global carbon dioxide emissions, the vast majority of which result from fossil fuel combustion. Transportation powered by biomethane has the potential to significantly reduce fossil fuel consumption and greenhouse gas emissions. Technically, fossil-fuelled natural gas could simply be substituted with biomass-derived biomethane, either liquefied or compressed.

The utilisation of biomethane in the transportation sector has several advantages: (1) biomethane is produced from organic biomass, which has nearly identical properties to natural gas; (2) biomethane produced by biogas plants can be produced and used locally, thereby avoiding the difficulties associated with natural gas importation, particularly in Europe (Eurostat 2022); and (3) employing biomethane in transportation systems would provide a carbon-saving option and effectively reduce climate change by 50–246% to gasoline- and diesel-fueled vehicles, depending on the calculations and well-to-wheel life cycles used, whereas fossil natural gas would reduce greenhouse gas emissions by 10–15% when compared to gasoline. Renewable liquefied biomethane is expected to significantly positively affect the climate. However, cost-effective small-scale liquefied biomethane schemes require further development.

The extensive use of inorganic fertilisers has resulted in risks to human health and the environment, such as a high carbon footprint. Digestate can assist in reducing the demand for mineral fertilisers by recovering nutrients from organic waste. As a result, digestate reduces our reliance on fossil natural gas, the need to transport these fertilisers globally, and all associated emissions. When one tonne of mineral fertiliser is replaced by one tonne of natural fertiliser, natural fertiliser could save one tonne of oil, 108 tonnes of water, and 5–9 tonnes of carbon dioxide equivalent emissions.

Some recommendations and prospects are obtained

- Biogas should not be measured solely in terms of the cost of a kilowatt-hour or megajoule of heat but rather in terms of the overall beneficial outcomes for environmental services and wider human benefits.
- Carbon is the most effective way to quantify all anaerobic digestion's environmental benefits. Carbon trading schemes, carbon taxes, or emissions limits, all increase the carbon gain value. As a result, any policy aimed at decarbonisation will positively contribute to the development of anaerobic digestion, and anaerobic digestion will positively contribute to decarbonisation.
- Incentives should be used to encourage sustainable agriculture, particularly in terms of carbon emissions reduction.

- Large-scale liquefied biomethane production should be subsidised to offset fossil fuel consumption and meet environmental standards.

Biogas upgrading

Biogas has the potential to be classified as a carbon sink renewable energy source because biogas derives the carbon from organic sources (livestock manure, sludge, food wastes, household organic waste, and agricultural residues), thereby lowering atmospheric carbon dioxide levels (Esposito et al. 2019). The biogas produced by organic biomass digestion was primarily composed of methane (40–75%) and carbon dioxide (15–60%), with a small amount of trace gases such as hydrogen sulphide (0.005–2%), nitrogen (0–2%), water vapour (5–10%), and other traces (Kapoor et al. 2019; Mahmudul et al. 2021).

Methane is the bioenergy-rich element in biogas, whereas other undesirable components reduce the sophisticated biogas utilisation as follows: (1) The presence of biogas contaminants, particularly the carbon dioxide, reduces the calorific value of biogas (around 20–25 MJ/cubic metre compared to 30–35 MJ/cubic metre of the natural gas), and substantially reduces the gas cylinder's volumetric capacity, thereby allowing the generated biogas to be used for low energy utilisation applications, e.g. cooking (Golmakani et al. 2022; Muñoz et al. 2015; Angelidaki et al. 2019). (2) Due to the limited infrastructure within biogas facilities, biogas is typically stored for a short period of time (a few hours), and as a result, biogas is typically stored under high pressure, a condition that results in the transformation of carbon dioxide, if present, obstructing biogas transportation (Kapoor et al. 2019; Angelidaki et al. 2018). (3) The presence of destructive hydrogen sulphide in raw biogas may cause significant damage to gas engines through sulphur dioxide emissions following ignition (Angelidaki et al. 2018). Thus, sequestering or removing carbon dioxide from raw biogas would reduce carbon dioxide emissions from anaerobic digestion systems, contribute to greenhouse gas mitigation, and have a greater positive environmental impact.

In many countries, households use raw biogas for heating and cooking without or with little monitoring of impurity effects. However, a more beneficial application of biogas is in the generation of electricity, transportation biofuel, natural gas substitute, and substrate for the chemical industry (Khan et al. 2021; Nguyen et al. 2021), and solid oxide fuel cells (SOFCs) or micro turbines applications (Saadabadi et al. 2019), which achieved via upgrading of biogas to biomethane. Due to the technical and environmental risks associated with impurities, several countries have proposed regulations governing biogas injection into natural gas grids, with a minimum methane content of more than 95% in many

cases (Angelidaki et al. 2019). The European biomethane standards for transportation fuel and grid injection require hydrogen sulphide concentrations to be less than 1 part per million and siloxanes concentrations to be less than 0.5 mg/normal cubic metre for grid injection and transportation fuel (Nguyen et al. 2021).

Numerous concise definitions exist for the biogas purification process: (1) biogas cleaning refers to the removal of other undesirable impurities (hydrogen sulphide, siloxanes, and ammonia, among others) that are harmful to gas grids or end-appliances; hydrogen sulphide cleaning is sometimes referred to as biogas desulphurisation; and (2) biogas upgrading refers to the removal of carbon dioxide from raw biogas in order to maximise the biomethane value of the final yield gas or the conversion of carbon (Angelidaki et al. 2019). After biogas upgrading, the final product is referred to as biomethane, which contains 95–99% methane. Germany and Sweden have the world's largest markets for biomethane. Other countries, particularly the United Kingdom, Switzerland, and France, are paying increasing attention (Nguyen et al. 2021).

The global market for biomethane was valued at $0.62 billion in 2017, and with a 26% annual growth rate, the market is expected to reach $4.96 billion by 2026. Numerous countries have established targets for replacing natural gas with biomethane in residential applications (Hoo et al. 2020). For instance, France intends to support 8 TW-h of biomethane production by 2023 (Herbes et al. 2018). The United Kingdom aims to use biomethane as the primary source of green gas in the future. On the other hand, the obtained biomethane can be compressed or liquefied to make biomethane more efficient in storage and transportation fuel. Liquefied biomethane is suitable for use in large vehicles, has a higher energy content, and has a long transport range. Recently, awareness of the benefits of liquified biomethane as a transportation fuel has grown. Sweden, for example, has set a target of using % biomethane-based transportation fuel by 2030.

Additionally, according to the European biomethane map, the total number of biomethane plants in Europe has doubled from 483 in 2018 to 729 in 2020. At the moment, 18 European countries produce biomethane. Germany has the most biomethane plants with 232, followed by France with 131 and the United Kingdom with 80 (EBA 2020). Around 47% of active biomethane plants in Europe are connected to the network grid, while 20% are connected to the transport grid. 10% of European biomethane plants are not connected to the grid, and information on the remaining 23% of plants is unavailable (EBA 2020). Figure 3 summarises the number of biomethane plants. In addition to the biogas purifications to meet the end-uses requirements, the removal and utilisation or storage of carbon dioxide can potentially decarbonise biogas or make biogas a carbon-negative energy resource.
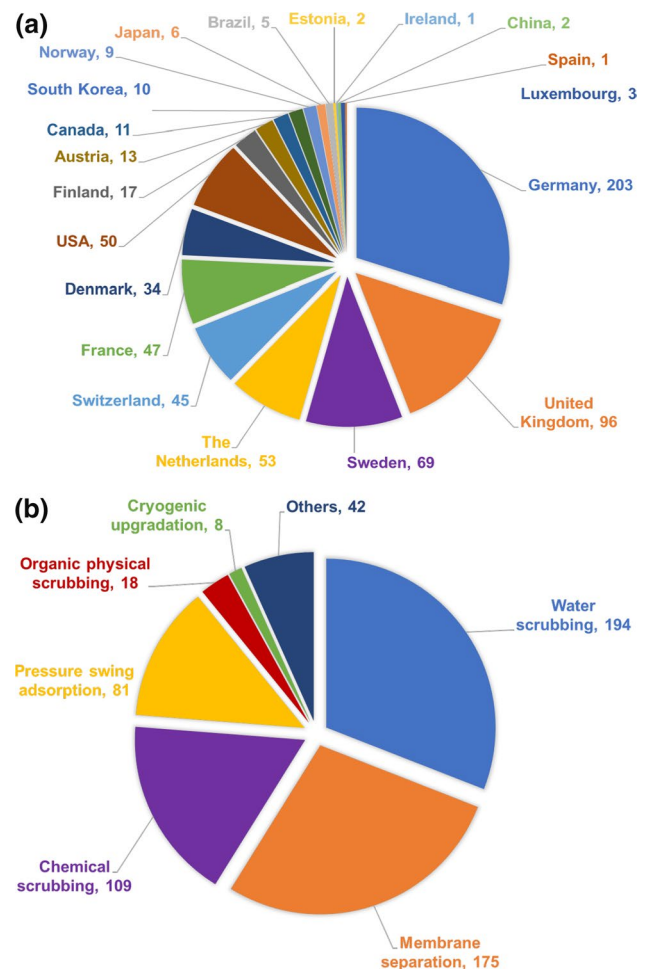
Fig. 3 Biogas upgrading plants and commercially available technologies (Bioenergy 2020). According to the European biomethane map in 2020, there are about 677 biomethane plants. Germany is the leading country with 203 biomethane plants, followed by the United Kingdom (96) and Sweden (69), as shown in **a**. Around 29% of the biomethane plants currently rely on water scrapping, 26% on membrane separation, and 16% on chemical scrubbing (**b**). Almost, biomethanation plants are present in Europe

Biogas upgrading technologies

Nowadays, several upgrading technologies aim to remove unwanted carbon dioxide from biogas to broaden the application range of biogas. These physical/chemical technologies, including water, organic, and chemical scrubbing, membrane separation, pressure swing adsorption, cryogenic separation, and biological-based methods, are widely used commercially. Scrubbers for water/chemicals, membrane separation, and pressure swing are well-established and widely used, accounting for more than 74% of the total market (Fig. 3), while in recent years, the biological biogas upgrading method has been proposed (Fu et al. 2021). The optimal approach for upgrading raw biogas depends entirely

on the biogas end-use, the efficiency of the upgrade, and the economics involved.

Scrubbing technologies

Water scrubbing has traditionally been based on the differences in the solubility of carbon dioxide and methane gas in the washing solution. Water (water scrubbing) or an organic solvent such as polyethylene glycol dimethyl ether (organic scrubbing) may be used as the washing solution. Carbon dioxide is 26 times more soluble in water than methane at 25 °C. Scrubbing methods frequently lack chemical reactions (physical process). Since gas solubility increases as pressure increases, pretreated biogas can be pressurised and introduced into the scrubbing column (Nguyen et al. 2021).

Hydrogen sulphide is first removed from raw biogas using a scrubber in the water scrubber method. The desulphurised biogas is then compressed to approximately 4–6.5 bar and delivered from the bottom side of the washing column to meet the injected water from the top. Carbon dioxide is absorbed by water, while biomethane diffuses from the top of the washing column. After drying the biomethane, biomethane is purified using an activated carbon filter to remove volatile organic carbon and then compressed into the gas grid (Ardolino et al. 2021). The carbon dioxide-rich water wash is injected into a stripping column, which uses atmospheric pressure air to remove carbon dioxide from the water (i.e. water reusable) (Nguyen et al. 2021). Water scrubbing technology is more than 98% effective at removing carbon dioxide (Angelidaki et al. 2018).

Organic scrubbing is similar to water scrubbing, but organic scrubbing purifies raw biogas using organic solvents such as propanol and polyethylene glycol. This is a more efficient process than water scrubbing because carbon dioxide dissolves more readily in an organic solvent than in water. This process yields 93–98% biomethane (Nguyen et al. 2021; Mulu et al. 2021; Singhal et al. 2017).

Chemical scrubbing is typically carried out with solvents derived from common organic amines such as methyldiethanolamine, diethanolamine, monoethanolamine, and diglycolamine (Nguyen et al. 2021). In principle, the operation is similar to that of water scrubbing; however, amine solvents have a higher absorption efficiency for carbon dioxide than water per unit volume, making them more effective at removing larger amounts of carbon dioxide and thus requiring smaller upgrading units (Ardolino et al. 2021). Due to the fact that the chemical adsorbent method is only reactive with carbon dioxide, methane leakage is minimal (0.1–0.2%) (Sun et al. 2015). As a result, post-combustion of the lean gas is unnecessary. Additionally, chemical scrubbing can produce methane with a purity of 99%. However, hydrogen sulphide must be removed upstream to avoid a degradative and corrosive reaction with the amine solution (Nguyen et al. 2021).

Pressure swing adsorption

This method utilises a porous medium to adsorb the target molecule from an injected gas; the adsorbed molecule is then released using a high-pressure value (Ntiamoah et al. 2016). By considering the various molecular dimensions of methane (0.38 nm) and carbon dioxide (0.34 nm), pressure swing adsorption units can be used to upgrade biogas (Ardolino et al. 2021). Consequently, filling an adsorbent substrate, such as activated carbons and zeolites with pore sizes of 0.37 nm in a column can retain carbon dioxide within the pores, while methane flows freely without being retained (Nguyen et al. 2021; Ardolino et al. 2021). Hydrogen sulphide removal from the upstream biogas is required in this method, as hydrogen sulphide is permanently removed by the adsorption material and has toxic effects.

The pressure swing method is accomplished by compressing the pretreated raw biogas to 4–8 bar and then injecting biogas into the sorption column via a bottom inlet. Thus, the smaller carbon dioxide molecules accumulate on the adsorption surfaces or are retained in the pores, whereas the larger methane molecules remain primarily in the gas phase and exit the column top as a biomethane-rich byproduct gas. Once the methane is released, the column pressure decreases to atmospheric pressure, releasing adsorbed carbon dioxide from the material surfaces and converting carbon dioxide to gas. The carbon dioxide-rich exhaust gas is vented through a valve located at the column's base. The column is then injected with biogas to initiate a new cycle of upgrading (Nguyen et al. 2021).

Membrane separation technology

The membrane separation technique is based on the theory that biogas permeates through a different membrane pores selectivity, where the membrane is highly permeable (20 times more permeable) to a small molecule (carbon dioxide) and impermeable to a large molecule (methane) (Nguyen et al. 2021). The permeated carbon dioxide is extremely pure (99.9%, particularly at −30 °C for separation of oxygen, nitrogen, and residual methane) and can be used in the beverage and food industries (Esposito et al. 2019), or carbon dioxide can be liquefied or compressed for use in other ways (Nguyen et al. 2021).

Membrane permeation occurs in various designs, with operating pressures ranging from 7 to 36 bars (Kapoor et al. 2019; Peppers et al. 2019). The membrane can be designed in two-stage cascades to achieve high methane purity. Where the gas can be returned from the first membrane to the inlet

via a circulation loop, while the methane-enriched gas flows into the second membrane (Nguyen et al. 2021).

Cryogenic treatment

The principle of cryogenic technology stems from the fact that gases condense differently at high pressure or low temperature. Carbon dioxide can solidify at $-78.5\ °C$ and 1 bar, whereas methane remains gaseous. As a result, gaseous methane can be easily separated from solidified carbon dioxide. Cryogenic treatment can achieve up to 99.9% methane or carbon dioxide purity with less than 1% methane loss.

However, the increased energy demand for refrigeration and compression of the gas is the primary constraint to this technology, as the required energy consumption accounts for 10% of the generated methane. Additionally, preventing frozen carbon dioxide from clogging the equipment and removing biogas impurities are critical issues (Nguyen et al. 2021); thus, this technology is not yet been fully established (Nguyen et al. 2021). Utilising biomethane as a liquefied biomethane ($-125\ °C$ and 15 bar), and selling frozen carbon dioxide as dry ice, can help this technology consume less energy and overcome some of the biomethane limitations (Esposito et al. 2019), thereby increasing biomethane commercial viability.

Overall, the operational requirements of different biogas upgrading methods are listed in Table 5.

Drawbacks of traditional biogas upgrading methods

Traditional biogas upgrading technologies are widely used and account for approximately 99% of all biogas upgrading plants (Khan et al. 2021). Nonetheless, these technologies have a number of limitations that may result in an increase in the cost of upgrading raw biogas. For instance, a water scrubber, which is frequently used in 41% of biogas upgrading plants, consumes enormous amounts of water (each 1000 normal cubic metre/hour gas flow consumes 200 cubic metres/hour of water); thus, regenerating water significantly increases the cost of water treatment (Sun et al. 2015; Hoyer et al. 2016). Additionally, the water scrubbing technique can result in up to 3% or more methane loss, and exhaust gas combustion is required for emission regulation (Ardolino et al. 2021). Water scrubbing may be economically viable when applied at wastewater treatment plants where effluents from the secondary and tertiary treatment stages are used as non-regenerating water resources (Angelidaki et al. 2018).

Amine absorption is an energy-intensive technology than physical scrubbing due to the regeneration of an amine solution that strongly binds with the gas molecules (Hosseini-ipour and Mehrpooya 2019), which increases the running costs of the process. The carbon dioxide-saturated amine solution must be heated to more than $120\ °C$ for regeneration, and then the remaining amine solution is cooled to $40\ °C$ prior to initiating a new upgrading cycle. The regeneration process mostly utilises 0.4–0.8 kW-h/normal cubic metre of biogas, nearly 15–30% of the energy produced from the generated biomethane (Angelidaki et al. 2018). Equipment corrosion, amine degradation, and volatile substance releases into the atmosphere are other drawbacks of the chemical scrubbing upgrading technology (Meng et al. 2019). Additionally, amines can degrade into nitramines and nitrosamines, which can have a detrimental effect on humans and the environment. Thus, the toxicity of solvents to humans and the environment, the high energy required to regenerate chemicals, the higher primary cost of amine solutions, and their evaporation loss are the primary disadvantages of chemical scrubbing (Angelidaki et al. 2018; Nguyen et al. 2021).

Organic scrubbing is an expensive and energy-intensive process. Additionally, organic solvent regeneration is more complicated than water regeneration. Pressure releases and air stripping are ineffective methods of organic solution regeneration. In practice, organic solvent regeneration requires an additional energy input of 0.1–0.15 kW-h/normal cubic metre of biogas from the solvent heating to $40–80\ °C$. This process can upgrade biomethane to a concentration of 93–98% (Nguyen et al. 2021; Mulu et al. 2021; Singhal et al. 2017).

The primary disadvantage of swing adsorption and membrane technologies are increased costs (Khan et al. 2021). In general, the advantages and disadvantages of conventional biogas upgrading methods are listed in Table 6.

In terms of emissions, the carbon dioxide removed from biogas during many conventional upgrading processes is frequently released into the environment, which not only emits the greenhouse gas but also wastes a valuable byproduct that could be converted into other chemicals (Golmakani et al. 2022; Zhu et al. 2019). As a result, additional technologies to benefit biogas and biogas byproducts must be developed to ensure that no waste is released into the environment.

Golmakani et al. (2022) estimated the global warming potential of the conventional biogas upgrading techniques at carbon dioxide and methane to be 1 and 28 times, respectively, without accounting for the energy-consuming in the separation process. The authors reported that the highest global warming potential is caused by membrane separation if not flared due to higher methane loss. Nevertheless, membrane separation had the lowest global warming potential if the waste methane was flared. The authors added that, given the high energy consumption associated with the separation process, the 1–2% methane loss due to water scrubbing could represent a real loss of 17–18%. Bakkaloglu et al. (2021) estimated the amount of methane emitted by ten

Table 5 Operational requirements of different biogas upgradation methods (Golmakani et al. 2022; Kapoor et al. 2019; Angelidaki et al. 2018; Khan et al. 2017; Sahota et al. 2018a)

Upgrading method	Operation theory		Operation temperature (°C)	Hydrogen sulphide cleaning requirement	Running pressure (bar)	Solvent or adsorbent agent	Heat required	Energy consumed/ upgraded biogas (kilowatt-hour/nor-mal cubic metre)	Methane purity (%)	Methane loss (%)	Investment cost (€/nor-mal cubic metre biogas)	Maintenance costs (€/year)	Operation simplicity
Water scrubber	Physical sorption		20–40	No	6–10	Water, anti-fouling agents	No	0.2–0.5	95–98	1–3	0.13–15	15,000	Simplest
Organic scrubber	Physical sorption		10–20	No	4–7	Organic solvents	70–80 °C	0.10–0.33	<93–98	1.5–4	0.25	39,000	Difficult in operation
Chemical scrubber	Chemical absorption		35–50	Required	1	Amines solutions, antifouling agents	120–160 °C	0.05–0.25	>98	0.04–0.1	0.28	59,000	Difficult in operation
Pressure swing adsorption	Adsorption		5–30	Required	4–8 bar	Adsorbents, molecular sieves	No	0.16–0.43	>96–98	1–3.5	0.26	56,000	Extensive process control needed
Membrane separation	Gas–Gas	Permeation	25–60	Required	20–36	Membranes of silicone cellulose acetate, rubbers, hollow fibres	No	0.18–0.35	90–92 96	0.5–20	0.22	25,000	Easy
	Gas–Liquid	Absorption											
Cryogenic separation	Multistage compression and condensation		−59 to −45	Recommended	40	Glycol	No	0.2–0.79	99	0.5–3	0.40–0.44	Not mentioned	Complex, multifaceted

Water scrubbing represents the lowest investment technology, while cryogenic separation is the most expensive technology. Chemical scrubbing and organic scrubbing require heating steps; however, the other methods do not require heating. Notably, pressure swing adsorption and high pH water scrubbing can eliminate carbon dioxide and hydrogen sulphide. However, pre-treatment to remove hydrogen sulphide is mandatory in amines scrubbing, pressure swing, and membrane separation techniques. All the technologies are energy-intensive

Table 6 Merits and drawbacks of biogas upgrading technologies (Golmakani et al. 2022; Fu et al. 2021; Qyyum et al. 2020)

Method	Advantages	Disadvantages
Water scrubber	Ideal for cold areas Most popular High methane purity and low loss Inexpensive Co-removal of hydrogen sulphide Does not need a special chemical or equipment Pre-cleaning is not required	Need high pressures of 6–20 bar and cooling units (< 20 °C) Form foams Biomethane drying is essential Hydrogen sulphide damages equipment Building up of elemental sulphur Environmental problems of acidification, global warming, and human toxicity are common Clogging due to bacterial growth High water demand
Organic scrubber	Higher carbon dioxide solubility than water scrubbing Lower operating and capital costs Remove hydrogen sulphide and other impurities Lower regeneration temperature (40 °C) Relatively low methane loss	Expensive investment and operation Only inert or gas steam (not air) should be used for high hydrogen sulphide regeneration Complex Regeneration required heating Requirement of chemicals
Chemical scrubber	Highest biomethane purity Lowest methane loss High carbon dioxide elimination efficiency Low gas pressure required in the absorption column reduces capital costs	High cost High energy consumption for regeneration Equipment corrosion Foaming Salt precipitation Hydrogen sulphide poisoning
Pressure swing	Does not enquire chemicals and heat Compact	High methane loss Complicated Pre-treatment is required High investment costs
Membrane separation	Compact and simple construction Commercial application Easy maintenance and operation High flexible shapes and mechanical stability Does not enquire chemical or heat Reliable Low energy consumption	Requires multiple processes for high purity methane High to medium methane losses High-pressure requirement (20–36 bar) Physical ageing Expensive Fouling and clogging of membranes Requires membrane replacement every 1–5 years Requires pre-treatment to remove hydrogen sulphide, water, siloxanes, and ammonia Energy and capital intensive Long-term instable
Cryogenic separation	High methane purity Removal of all impurities	Highest energy consumption (10% of the generated methane) High losses of methane Practical problems (e.g. clogging)

No ideal biogas upgrading method has existed. Almost the barriers facing these technologies originated from an economic and technical point of view. In addition, the conventional biogas upgrading technologies usually require additional equipment to produce high-pressure or high temperatures conditions, which raises the cost of these technologies

biogas plants in the United Kingdom and discovered losses ranging from 0.02 to 8.1%.

Additionally, methane emissions from small-scale farm plants were higher than those from large-scale biogas plants that treated food waste. The authors concluded that biogas plant methane emissions could account for up to 1.9% of total methane emissions in the United Kingdom. Florio et al. (2019) reported that the global warming potential of the chemical scrubbing process, cryogenic process, membrane separation, and pressure swing adsorption was 1.27, 1.16, 1.09, and 1.11 kg carbon dioxide equivalents per cubic metre of used biogas, respectively. Similarly, Hauser (2017)

showed that the biogas upgrading of cryogenic technology had a lower climate change impact of 183, 83, 71, and 73% than those of membrane technology, pressure swing, amine scrubbing, and high-pressure swing separation, respectively, through life cycle assessment (Hauser 2017).

To summarise, all commercially available biogas upgrading processes contribute to global warming to varying degrees. As a result, searching for a more environmentally friendly method is critical.

Hydrogen-assisted biogas upgrading technology

Along with the economic barrier, increased concern about the environmental consequences of conventional biogas upgrading methods is confirmed (Golmakani et al. 2022); thus, adopting more environmentally sustainable alternatives is required. In this context, biological biogas upgrading has garnered considerable interest as a viable technology for increasing the methane value of anaerobically generated biogas while simultaneously stabilising waste biomass (Treu et al. 2018). The biological biogas upgrading technology is a hydrogen-assisted process that proposes to capture carbon dioxide by converting carbon dioxide to biomethane via the use of hydrogen generated primarily from other renewable energy sources such as water electrolysis, photovoltaic solar facilities, or wind turbines, thereby forming a new energy shifting technology called power to gas shift (Fu et al. 2021; Zhu et al. 2020a; Zabranska and Pokorna 2018). The energy generated by solar and wind renewable energy sources is stored in biomethane, with hydrogen acting as an intermediary energy carrier (Omar et al. 2019; Bassani et al. 2017).

Wind and solar energy generation are renewable, sustainable, and clean energy source that has gained recent attention. Recently, the cost of renewable-energy-based electricity generation has decreased significantly. Solar energy costs have decreased from \$0.378/kW-h in 2010 to \$0.068/kW-h in 2020, a savings of 82%, and are expected to continue decreasing to approximately \$0.02/kilowatt-hour in 2050 (Lai et al. 2021). In Germany, the renewable energy share in the net electricity production increased from 40.6% in 2018 to 46% in 2019, surpassing fossil fuels for the first time, with 24.6 and 9.0% of renewable energy generated from wind and solar, respectively (ISE 2020). Renewable energy accounts for 36.9% of electricity generation in the United Kingdom, with approximately 20% from wind (BEIS 2022). Solar and wind energy accounted for 49% of Danish electricity consumption in 2019 (State-of-Green 2020).

Effective electricity storage and utilisation are critical; otherwise, excess energy can easily be lost, and power grids become unstable. Batteries can be used to store electricity; however, they have a limited storage capacity, are expensive, and pose significant environmental hazards due to their obsolete constituents. Wind and solar energy conversion to hydrogen are attractive because hydrogen is a form of clean energy produced through water electrolysis (Osman et al. 2022). While hydrogen has a lower energy density (10.88 MJ/cubic metre), the challenges are associated with hydrogen storage and use as a transportation fuel (Lai et al. 2021). By contrast, biomethane has a much higher energy density (36 MJ/cubic metre) than hydrogen, is compatible with active storage infrastructure such as the gas grid, and
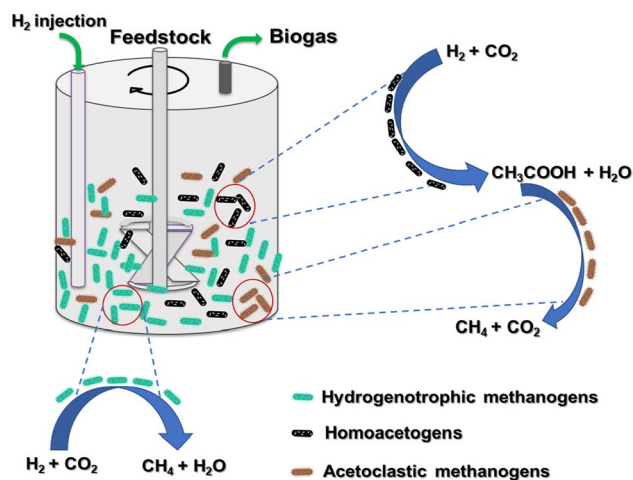
Fig. 4 Different microbial pathways exist for biogas upgrading via hydrogen injection. Hydrogen injection into the anaerobic digestion bioreactor facilitated the hydrogenotrophic methanogens' utilisation of carbon dioxide to produce biomethane. Additionally, the Homoacetogens can convert carbon dioxide to acetate, which is primarily consumed by acetoclastic methanogens in methane production. Syntrophic relationships between bacteria and archaea are critical for system stability

has a broad range of applications, including electricity and vehicle fuel. As a result, hydrogen-driven chemoautotrophic biogas upgrading technology demonstrates an exceptional ability to convert intermittent energy sources (solar and wind) to more stable energy sources (biomethane) that can be easily stored, thereby fostering the development of an environmentally friendly, sustainable, and circular economy shifting concept (Lai et al. 2021).

Three distinct types of upgrading configurations for hydrogen-driven upgrading technology can be implemented: namely, in situ, ex-situ, and hybrid-situ systems.

Microbial process in chemoautotrophic carbon dioxide conversion using hydrogen

Methanogenic archaea are the primary metabolic microorganisms responsible for methanogenesis in the anaerobic digestion system. They do so by extensively utilising intermediates such as hydrogen, carbon dioxide, alcohols, and short-chain volatile fatty acids that result from the biodegradation of complex organic materials to produce methane (Wu et al. 2021). Three classes of methanogens, namely acetoclastic methanogens, hydrogenotrophic methanogens, and homoacetogens, predominate in the anaerobic digestion process based on the kind of substrates used for their metabolism, as indicated in Table 7 (Fu et al. 2021; Laiq Ur Rehman et al. 2019).

Table 7 Methanogens species associated with intermediates for methane generation (Wu et al. 2021)

Acetotrophic methanogens		Hydrogenotrophic methanogens		*Homoacetogens*	
Species	Substrate	Species	Substrate	Species	Substrate
Methanosarcina ace-tivorans	Methanol and acetic acid	*Methanobacterium bryantii*	Hydrogen and carbon dioxide	*Treponema*	Hydrogen and carbon dioxide
Methanosaeta concilii (soehngenii)	Acetic acid	*Methanobacterium ther-moalcaliphium*	Carbon dioxide and hydrogen	*Clostridium ljungdahlii*	Carbon dioxide and hydrogen
Methanosaeta ther-mophila	Acetic acid	*Methanothermobacter thermoautotrophicm*	Carbon dioxide and hydrogen	*Lysinibacillus fusiformis*	Carbon dioxide and hydrogen
		Methanothermovacter wolfeii	Carbon dioxide and hydrogen	*Bacillus cereus*	Carbon dioxide and hydrogen
		Methanolacinia paynteri	Carbon dioxide and hydrogen	*Lutispora*	Hydrogen and carbon dioxide
		Methanobacterium formicicum	Carbon dioxide; hydrogen; formic acid		
		Methanobrevibacter smithii	Carbon dioxide; hydrogen; formic acid		
		Methanosarcina barkeri	Hydrogen and carbon dioxide; acetic acid methylamine; methanol		
		Methanosarcina ther-mophile	Hydrogen and carbon dioxide; acetic acid methylamine; methanol		

Three classes of methanogens, namely acetoclastic methanogens, hydrogenotrophic methanogens, and *Homoacetogens*, prevail over the anaerobic digestion process based on the kind of substrates used for their metabolism. The acetoclastic methanogens usually utilise acetate, while hydrogenotrophic methanogens and *Homoacetogens* mostly consume hydrogen and carbon dioxide. Shifts in microbial dominance can be observed after hydrogen injection into the system, in which the hydrogenotrophic methanogens will be the dominant bacteria to utilise the hydrogen into methane
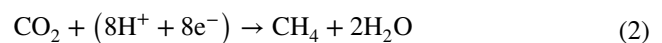
Hydrogenotrophic methanogens

Hydrogenotrophic methanogens are likely the microorganisms that perform methanogenesis by utilising hydrogen as a primary energy source in conjunction with other substrates to convert carbon dioxide to biomethane, as illustrated in Fig. 4. The methane produced from the carbon dioxide and hydrogen interactions can mitigate hydrogen's adverse effect on acetogenic methanogens, and this is a thermodynamically favourable reaction, as seen in Eq. 1, which is critical for the anaerobic digestion stability of the subsequent hydrogen-driven biogas upgrading technology (Zhu et al. 2020a). The methane produced in this reaction is combined with other gases in the biogas, resolving technical difficulties associated with the storage and transportation of hydrogen posed by the explosion risk potential (Zabranska and Pokorna 2018).

$$4H_2 + CO_2 \rightarrow CH_4 + 2H_2O \quad \Delta G^{\circ\prime} = -130.7 \text{ KJ/mol} \quad (1)$$
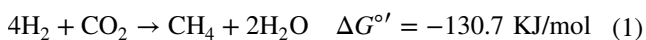
In addition to the hydrogen, some other organic intermediates such as acetic acid and methanol may be utilised as electron donors to convert carbon dioxide for methane generation by some hydrogenotrophic methanogens like *Methanosarcina* spp, as shown in Table 7 and Eq. 2 (Yee and Rotaru 2020).

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

A high ammonia nitrogen level of about 5500 mg/l was reported to stimulate favourable effects on transforming carbon dioxide into methane (94.1%) by hydrogenotrophic methanogens; in addition, they showed higher robustness (abundance 73.1%) to ammonia inhibition than acetotrophic methanogens (abundance 1.3%) (Li et al. 2020). Consequently, hydrogenotrophic methanogens could have a vital role in biogas upgradation under higher ammonia stress.

Increasing hydrogenotrophic methanogens abundance could also prevent the imbalance between hydrogen production and consumption, and hence increasing methane yield. In addition, the hydrogen inside the anaerobic digestion reaction has a critical role in interspecies electron exchange between syntrophic bacteria and methanogens. The interspecies electrons transmission might boost the microbial communities' multiplicity, hence producing more energy from interactions that cannot be catalysed by a single microbe (Shrestha and Rotaru 2014). This indicates

that hydrogen injection has various effects on the anaerobic digestion process, and the effect of hydrogen on interspecies electron exchange necessitates additional investigation (Fu et al. 2021).

Homoacetogens

Homoacetogen microorganisms can convert carbon dioxide into acetate via the Wood-Ljungdahl pathway; the acetates are then used to produce methane by acetoclastic methanogens (Fig. 4), hence upgrading biogas indirectly (Angelidaki et al. 2018) (Eq. 3).

$$4H_2 + 2CO_2 \rightarrow CH_3COOH + 2H_2O \quad \Delta G^{\circ\prime} = -104.5 \text{ KJ/mol} \tag{3}$$
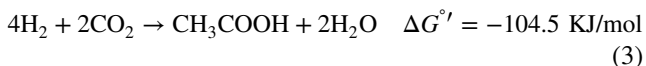
Homoacetogen favours low temperature (psychrophilic) for better proliferation when hydrogen is sufficient than hydrogenotrophic organisms (Braga Nan et al. 2020). Hence, the *Homoacetogens'* role in hydrogen consumption is insignificant under the thermophilic rector and when the hydrogen is inadequate. Higher hydrogen pressure could change the metabolic pathway inside the anaerobic system towards *Homoacetogens* while suppressing methanogenesis (Wu et al. 2021). Notably, increased partial exogenous hydrogen pressure led to shared hydrogen consumption of 60% and 40% for hydrogenotrophic methanogens and *Homoacetogens*, respectively, to utilise up to 40% of externally injected hydrogen (Liu et al. 2016). Thus, indirectly converting hydrogen to acetate and then to methane via *Homoacetogens* could increase the calorific value of ultimate biogas produced following exogenous hydrogen injection into anaerobic digestion systems.

Acetoclastic methanogens Acetoclastic (acetotrophic) methanogens can convert acetate to methane and carbon dioxide (Fig. 4); thus, the performance and the bioactivity of acetoclastic methanogens are critical throughout the anaerobic conversion of acetate, as shown in Eq. 4.

$$CH_3COOH \rightarrow CH_4 + CO_2 \quad \Delta G^{\circ\prime} = -31 \text{ KJ/mol} \tag{4}$$

Methanosarcina and *Methanosaeta* are the most abundant acetotrophic methanogens in the anaerobic digestion operation. The availability of the feedstock and the operating conditions can influence the dominance of the two methanogens. Low acetate levels favoured the dominance of *Methanosaeta*, whereas increased ammonia and volatile organic acid levels favoured *Methanosarcina* dominance (Zabranska and Pokorna 2018). Overall, filamentous acetoclastic methanogens are less resistant to hydrogen sulphide, ammonia, and volatile fatty acids concentrations than hydrogenotrophic methanogens or the *Methanosarcinaceae* (Zabranska and Pokorna 2018). *Methanosarcina*, Acetotrophic methanogens,

was predominant in food waste digesters, while *Methanosaeta* was dominant in wastewater treatment digesters (Kim et al. 2019).

Configurations of hydrogen-based biogas upgrading

The biogas upgrading system can be classified into in situ, ex-situ, or hybrid designs depending on how hydrogen is fed to the anaerobic digester to capture carbon dioxide and convert carbon dioxide into biomethane, as seen in Fig. 5.

In situ hydrogen delivery The in situ hydrogen delivery is a simple system that operates with one water electrolysis unit for hydrogen generation and one anaerobic reactor, as shown in Fig. 5. Specifically, hydrogen is fed directly into anaerobic digesters to aid in converting endogenous carbon dioxide produced during anaerobic digestion to methane; simultaneously, the delivered hydrogen stimulates the activity of hydrogenotrophic methanogens (Lai et al. 2021).

The in situ upgrading systems have the ability to treat several organic feedstocks, such as manure (Zhu et al. 2019b) and food waste (Kim et al. 2021), as listed in Table 8. In this upgrading method, methane content can typically be increased from > 68% to up to 100%. Kim et al. (2021) demonstrated that in situ delivering hydrogen to an anaerobic digester treating food wastes increased the hydrogenotrophic methanogens like *Methanobacterium*, *Methanosarcina*, and *Methanosaeta* in the system. Additionally, operation
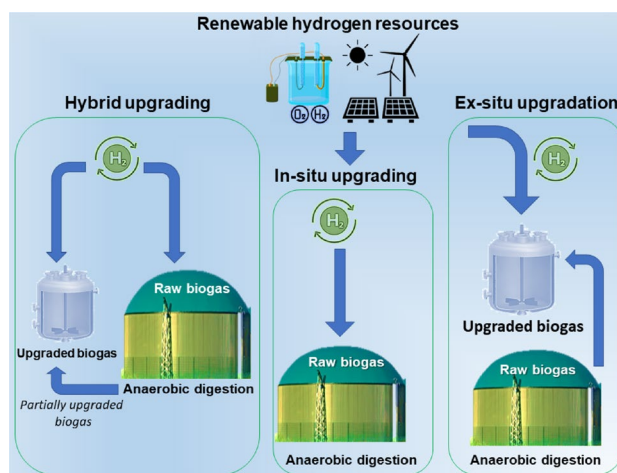
Fig. 5 The various configurations used for hydrogen-assisted biogas upgrading processes include in situ hydrogen injection directly into the reactor, ex-situ hydrogen injection into a separate hydrogen-containing unit and hybrid processes that combine the two systems. The delivered hydrogen stimulates the activity of hydrogenotrophic methanogens to utilise and convert endogenous carbon dioxide into biomethane. The in situ hydrogen delivery systems face several challenges, such as low hydrogen solubility, low hydrogen utilisation rate, and increased system pH. However, the ex-situ hydrogen delivery shows comparatively low drawbacks, whereas the hybrid processes in under development

Table 8 Performance and upgrading abilities of in situ biogas systems

Reactor volume (L)	Operation temperature (°C)	Feedstock	pH	Methane generation rate (litre/litre$_{reactor}$/day)	Hydrogen utilisation efficiency	Methane content (%)	References
3.0 (continuous stirred reactor)	37	Food waste	7	0.73	96%	92	Kim et al. (2021
35	35	Sewage sludge	7	0.59	99%	95.2	Díaz et al., 2020
2 (continuous stirred reactor)	38	Sludge and straw	7.9	0.34–0.44	58–99%	77–100	Agneessens et al., 2017
0.075	37	Food waste	8.5	0.09	72%	77	Okoro-Shekwaga et al., 2019
11.6 (continuous stirred reactor)	35	Swine manure	7.59	0.44	22%	70	Zhu et al., 2020b
2.0	38	Sludge and straw	7.9	0.44	100%	100	Agneessens et al., 2017
11.2	35	Swine manure	7.4–7.59	Not mentioned	0.9–1.9 L/day	65–70	Zhu et al. (2019b)
11.2	55	Swine manure	7.64–7.77	Not mentioned	1.9–6.4 L/day	68–78	Zhu et al. (2019b)
11.2	55	Sodium formate & pig manure	7	Not mentioned	1.9–6.6 L/day	68–83	Zhu et al. (2019a)
Headspace of 1.4 L and actual liquid volume of 0.7 L (two-stage up flow anaerobic sludge blanket reactor)	35	Synthetic wastewater, glucose	7	Not mentioned	< 99%	> 90	Xu et al. (2020)
1.4	55	Potato-starch wastewater	8.4	1.15	94%	81	Bassani et al. (2016)
6.3 (up-flow anaerobic sludge blanket reactor)	37 55	Wastewater sludge	7.2	0.4–4.8	< 1.6 L/litre$_{reactor}$/day	96	Yun et al. (2017)
0.12	52	Maize leaf	7–8	0.13	100%	89	Mulat et al. (2017)

The in situ biogas upgrading can consume 58–100% of injected hydrogen gas to produce 68–100% biomethane under different operating temperatures and with various feedstocks. The daily methane production rate varies with an optimum of 4.8 L/litre$_{reactor}$/day. The different rates of biomethane are attributed to the different reactor configurations. The in situ biogas upgrading still lacks large-scale system application to emphasise the potential role of hydrogen injected on the biomethanation of endogenously produced carbon dioxides

temperature was critical for hydrogen utilisation rate and domaining microbial communities in the in situ upgrading digesters (Zhu et al. 2019b). Whereby, in the mesophilic digester (35 °C), almost all hydrogen was consumed by acetotrophic methanogens and *Homoacetogens*. However, at thermophilic operations (55 °C), hydrogen was mainly used to maintain cell growth, with a fraction of hydrogen being used by hydrogenotrophic/acetotrophic methanogens (Zhu et al. 2019b).

As previously mentioned, there are two main methanogenesis pathways: acetotrophic methanogenesis (Eq. 4) and hydrogenotrophic methanogens (Eq. 1). The hydrogenotrophic methanogens are more thermodynamically favourable and stable than the acetoclastic methanogenesis (Sarker et al. 2018). Hydrogenotrophic methanogens have been investigated in pilot-scale biodigesters for biogas upgradation (Dupnock and Deshusses 2021). Theoretically, hydrogenotrophic methanogens could utilise 4 mol of hydrogen and 1 mol of carbon dioxide as electron donor and acceptor, respectively, to generate 1 mol of methane (Eq. 1) (Zhao et al. 2021).

Besides, the carbon dioxide is converted into acetate, catalysed by *Homoacetogens* via Eq. 3, and then acetotrophic methanogens utilise acetates to generate biomethane (Eq. 4) (Fu et al. 2021). Notably, hydrogenotrophic methanogens are extensively dispersed within the methanogens, where *Methanosarcina*, *Methanobacterium*, and *Methanoculleus* could use the hydrogenotrophic methanogens pathway for methane production, and previously mentioned, where the hydrogenotrophic methanogens are the most dominant archaea in in situ hydrogen supplemented systems (Zhao et al. 2021). Therefore, hydrogen feeding into the in situ

reactor shifts the methanogenic metabolic pathway towards the hydrogenotrophic methanogens pathway.

Ex-situ hydrogen-based biogas upgradation For the ex-situ biogas upgrading system, raw biogas produced from an anaerobic reactor is introduced into another anaerobic reactor with hydrogen to convert the carbon dioxide in the raw biogas into biomethane (Fu et al. 2021) via hydrogenotrophic methanogens pathway (Table 9 and Fig. 5). Additionally, hydrogen is obtained through the renewable water electrolysis process. Compared to in situ hydrogenotrophic methanogenesis, the external bioreactor requires only carbon dioxide, hydrogen, basic nutrients, and hydrogenotrophic cultures (Kougias et al. 2017; Das et al. 2022).

Bassani et al. (2015) combined two continuous stirred bioreactors to upgrade biogas; while the first bioreactor was used to generate biogas, while the second was used to upgrade biogas. When hydrogen was injected into the second bioreactor, bioreactors could produce biomethane with a purity greater than 85% in mesophilic and thermophilic operations. In addition, introducing hydrogen enhanced the growth of *Methanoculleus*, a hydrogenotrophic methanogen. In doing so, Kougias et al. (2017) attained methane purity of more than 98% using an ex-situ biogas bubble column upgrading bioreactor. A microbial assessment revealed that as well as hydrogenotrophic methanogens such as *Methanothermobacter* and *Methanoculleus*, as well as *Clostridia* spp., predominated.

Hybrid hydrogen-based biogas upgradation The hybrid concept couples in situ and ex-situ upgradation into a single process, in which carbon dioxide is converted to methane in the anaerobic digester followed by further carbon dioxide biomethanation in the ex-situ separate reactor, as illustrated in Fig. 5.

For example, Corbellini et al. (2018) used a biogas hybrid upgrading system comprising two-stage thermophilic upflow digesters. The authors injected hydrogen gas into the first digester, "in situ *upgrading*", which contained potato starch and cattle manure as feedstocks. The biogas generated from the in situ *upgrading* reactor was received in an ex-situ reactor, in which abundant hydrogenotrophic methanogen cultures were inoculated for hydrogenation of remaining carbon dioxide into biomethane. They found that biomethane with a rate of 0.355 L/l$_{reactor}$/day and 95% methane content was generated with a 96.5% hydrogen utilisation rate. Besides, carbon dioxide utilisation by hydrogenotrophic methanogens in the ex-situ bioreactor buffered the pH drop caused by the generation of organic acids in the first bioreactor. Moreover, the anaerobic microbiome significantly increases the diversity of hydrogenotrophic communities in the ex-situ bioreactor during hydrogen injection.

Biogas upgrading using Homoacetogens

Homoacetogens can be used to convert carbon dioxide in biogas into valuable substrates while also upgrading raw biogas to biomethane, and this approach has recently gained attention. Numerous *Homoacetogens*, such as *Acetobacterium woodii* (Cheng et al. 2018), *C. autoethanogenum*(Heffernan et al. 2020), and *Clostridium scatologenes* (Liu et al. 2018), can produce byproducts, such as acetate, ethanol, and butyrate using carbon dioxide when carbon dioxide is used as a carbon source, and hydrogen serves as the electron donor (Table 10). Compared to methane, these liquid biofuels generate more energy and have a higher market value, making them more suitable for transportation and storage.

Zhao et al. (2020) employed a membrane reactor inoculated with *Homoacetogen C. ragsdalei* P11 for biogas upgradation and biofuel generation. They generated biomethane with a content > 97% and acetic acid and ethanol generation rates of 37.8 and 13.5 mmol litre/day, respectively. Other authors also upgraded carbon dioxide to alcohols or volatile fatty acids using *Acetoanaerobium noterae* or *Moorella* spp. as the main *Homoacetogen* microorganisms. (Omar et al. 2019; Omar et al. 2018). These findings suggest that a *Homoacetogen* augmentation is a viable option for biogas upgrading. Nonetheless, to extract end-products from reactor systems containing a mixture of organic and alcohol chemicals.

Main bottlenecks of current hydrogen-assisted upgrading technologies

Without additional infrastructure, the low-cost asset is advantageous for in situ biogas upgrading. However, the hydrogen partial pressure and the changes caused by carbon dioxide and volatile fatty acid accumulations are significant constraints resulting in decreased biomethane production. Hydrolysis, acidogenesis, acetogenesis, and methanogenesis are the four stages of anaerobic digestion. Organic particulates are hydrolysed to form precursors, which are then converted biochemically to form liquors and volatile fatty acids, such as acetic acid, formic acid, butyric acid, and propionic acid (acidogenesis). These monomers are then biofermented to form acetate, formate, hydrogen, and carbon dioxide (acetogenesis), which methanogens use to produce methane (methanogenesis) (Li et al. 2019). Following that, syntrophic interactions between functional microorganisms are critical for the stability and performance of anaerobic digestion.

The sudden injection of a large amount of hydrogen gas may result in an imbalance between syntrophic parties, affecting all four upstream phases. Exogenous hydrogen injection, for example, suppressed syntrophic bacterial

Table 9 Running performance and upgrading abilities of ex-situ systems

Bioreactor configuration	Working volume (L)	Operation temperature (°C)	Inoculum source/microorganism involved	Methane production rate (litre/litre_reactor/day)	Hydrogen utilisation (%)	Methane content (%)	pH	References
Fed-batch reactor	1	37	Anaerobic granular sludge	Not mentioned	100	77–91	8	Tang et al. (2021)
Fixed bed reactor filled with granular perlite and vermiculite shales	4	53–55	Slurry of methanogens	6.35	100	>90	6.6–7.0	Alitalo et al. (2015)
Continuous stirred reactor	1.2	52	Anaerobic digestate	Not mentioned	Not mentioned	<79	8.0	Kougias et al. (2017)
Bio-trickling filter reactor	Not mentioned	35	*Methanospirillum hungatei*	Not mentioned	99	98	7.2	Dupnock and Deshusses (2019)
Trickle-bed bioreactor	58	37	Groups from an anaerobic digester	2.52	>99.9	95	7.4–7.7	Rachbauer et al. (2016)
Bubble column reactor	1.2	52	Anaerobic digestate	Not mentioned	100	97–98	8.0	Kougias et al. (2017)
Two-stage continuous stirred reactor	1.5–2	35	Biogas plant's sludge	0.10	93	89	7.7–8.0	Bassani et al. (2015)
Two serial up-flow reactors	2.8	52	Anaerobic digestate	Not mentioned	100	>98	8.0	Kougias et al. (2017)
Up-flow reactors with aluminium oxide ceramic membrane gas diffuser	0.9	56	Hydrogenotrophic enrichment and degassed digestate	0.7	100	94	8.1	Bassani et al. (2017)
Up-flow reactors with alumina ceramic sponge and stainless-steel diffusers	0.9	55	Hydrogenotrophic enrichment and degassed digestate	0.80	100	96	8.0	Bassani et al. (2017)
Anaerobic three-phases system	88	37	Wastewater anaerobic sludge	1.49	Not mentioned	98	7.2–7.4	Burkhardt et al. (2015)

The hydrogenotrophic methanogens from anaerobic digestate play a critical role in ex-situ biogas upgrades to convert carbon dioxide into methane by consuming the injected hydrogen. The ex-situ biogas upgrading can consume 93–100% of injected hydrogen gas to produce 77–98% biomethane under different operating temperatures and with various feedstocks. Two- and three-stages reactors with bubble column reactors and bio-trickling filter reactors produce the highest biomethane of up to 98%. The ex-situ systems could be a promising means of biogas upgradation to substitute the conventional upgradation technologies

Table 10 Carbon dioxide conversion to biofuels using *Homoacetogens*

Feeding	Reactor configuration	Operation temperature (°C)	Working volume (L)	pH	Inoculum/ microorganisms	Product and generation rate (gram/litre/day)	Products (g/l)	References
Carbon dioxide + hydrogen	Anaerobic membrane bioreactor	37	2	7.0	*Clostridium ljungdahlii*	Acetate: 1.68	Not mentioned	Cheng et al. (2018)
Biogas	Hollow fibre anaerobic membrane bioreactor	30	0.2	7.0–7.5	*Clostridium ragsdalei* P11	Methane: 97.6%, ethanol: 0.62, acetate: 2.27,	1.0 ethanol; 2.7 acetate	Zhao et al. (2020)
Biogas	Bottle	55	0.1	8.6	*Moorella* sp.	Not mentioned	0.5 acetate; > 77% methane	Omar et al. (2019)
Biogas	Bottle	37	0.1	4.7–6	*Acetoanaerobium noterae*	Not mentioned	0.3 acetate; > 95% methane	Omar et al. (2018)

Homoacetogen augmentation is a proper option for biogas upgradation with acetate, ethanol, and methane products. The methane can be upgraded up to 95%, while producing acetate at a rate of 2.27 g/l/day is possible. However, extracting valuable chemicals mixed with other organics from reactor systems is challenging

activity for the degradation of volatile fatty acids and alcohols, as the acetogenesis step is thermodynamically unfavourable at high hydrogen dosage pressures (greater than 10 pascals) (Lai et al. 2021) with 5.82 pascals is the optimum (Zabranska and Pokorna 2018).

The accumulated volatile fatty acids such as butyrate, propionate, ethanol, and lactate could inhibit methanogens (Omar et al. 2019; Angelidaki et al. 2018; Li et al. 2021). Additionally, hydrogen addition may enhance *Homoacetogens*, promoting carbon dioxide conversion to acetate. If the rate of acetate consumption decreased faster than the rate of production, the entire anaerobic digestion process would be inhibited (Mulat et al. 2017).

Alcohols and volatile fatty acids accumulations in an anaerobic digester as a result of direct hydrogen addition could also cause acidification (Angelidaki et al. 2018); thus, the anaerobic digestion process's optimal synergistic interactions will have deviated from the pH range of 6.8–7.4 (Farghali et al. 2020). Thus, the rate of hydrogen addition is critical in reactors. The optimal hydrogen to carbon dioxide ratio is unknown at the moment. According to some studies, a hydrogen-to-carbon dioxide ratio of 2:1 is optimal for simultaneous biogas upgrading (Omar et al. 2019; Omar et al. 2018). In comparison, another researcher demonstrated that a 4:1 ratio is optimal for biogas upgrading (Wahid et al. 2019). Increases in the hydrogen to carbon dioxide ratio above 4:1 may deplete the carbon source for hydrogenotrophic methanogens, thereby inhibiting the function of methanogenesis (Luo and Angelidaki 2013). Additional evaluations with various hydrogen to carbon dioxide ratios, feedstocks, and inoculum sources should be conducted to determine the optimal injection ratio.

On the other hand, the methanogenic phase may result in alkaline conditions. According to Eq. 5, methanogens may use carbon dioxide to reduce H^+ in the liquid, thereby increasing the pH value (Sarker et al. 2018). Additionally, the conversion of carbon dioxide to biomethane via the exogenous hydrogen addition pathway (in situ upgrading) reduces the levels of endogenous carbon dioxides in the liquid, raising the pH above 8.5, which suppresses biological processes (Zabranska and Pokorna 2018). A pH value greater than the optimum value, i.e. 8.5 for methanogenesis, would inhibit the activity of hydrogenotrophic methanogens (Angelidaki et al. 2018).

$$H_2O + CO_2 \rightarrow HCO_3^- + H^+ \tag{5}$$

Generally, the in situ biogas upgrading is a favourable technology for enhancing methane content in biogas; however, maintaining a balance between syntrophic bacteria and methanogens is critical for optimising biomethane generation in conjunction with exogenous hydrogen addition.
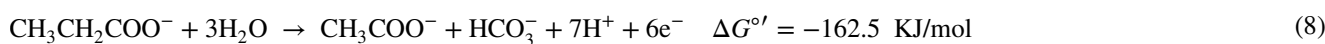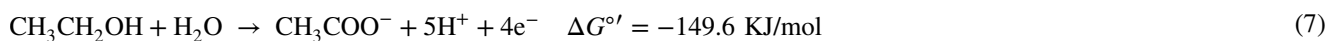
Lower hydrogen to liquid transfer rate One of the main limitations of the hydrogen-driven biogas upgrading process is the low hydrogen gas to liquid transfer ratio, which is initiated by the low hydrogen solubility in the liquid phase (0.76 mg/l at 80 °C and 1.93 mg/litre at 0 °C) (Rafrafi et al. 2021). Because biological biomethanation occurs primarily in the liquid phase, hydrogen must be transported across the line connecting the gas and the liquid in order for hydrogenotrophic methanogens to utilise hydrogen.

Consequently, the low hydrogen gas-to-liquid transfer rate is a limiting factor for hydrogenotrophic bioprocesses. Due

to the extremely limited hydrogen dispersion, this concern is critical for in situ upgrading. Despite the massive amounts of hydrogen injected during upgrading operations, hydrogen is only accessible to the methanogens' superficial layers (Zhao et al. 2021a). The efficiency of hydrogen utilisation is highly dependent on the rate of hydrogen gas to liquid transfer, as defined in Eq. 6.

$$H_{tr} = 22.4 L_g c \left(H_{2gP} - HL_{2l} \right) \tag{6}$$

where H_{tr} represents hydrogen transfer rate (mole/litre/hour), $L_g c$ is linear hydrogen transfer coefficient (*per hour*), H_{2gP} is hydrogen amount in the gas phase (mole/litre), and HL_{2l} is

$$CH_3CH_2OH + H_2O \rightarrow CH_3COO^- + 5H^+ + 4e^- \quad \Delta G^{\circ\prime} = -149.6 \text{ KJ/mol} \tag{7}$$

$$CH_3CH_2COO^- + 3H_2O \rightarrow CH_3COO^- + HCO_3^- + 7H^+ + 6e^- \quad \Delta G^{\circ\prime} = -162.5 \text{ KJ/mol} \tag{8}$$

the dissolved hydrogen in the liquid phase (mole/litre).

As a result, a suitable hydrogen diffusion device is required, as excess hydrogen easily leaks from the reactor. As a result, the low hydrogen utilisation efficiency would significantly increase the cost of hydrogen supply, reducing economic gains. Additionally, the escaped hydrogen could pose a threat to public safety due to the possibility of an explosion.

Low biomethane generation The generation rate of methane from in situ and ex-situ upgrading systems is mostly in the range of 0.1–1 and 0.1–3 L/l$_{reactor}$/day, respectively; in addition, the liquid (biofuel) generation rate via *Homoacetogens* is typically below 5 g/l/day for acetate (Lai et al. 2021). Accordingly, a higher generation rate is required to ensure the economic viability and technical feasibility of upgrading biological biogas for industrial purposes.

Prospective barriers solutions

Reducing in situ metabolic limitation Pulsed hydrogen injection may be a viable option for alleviating in situ metabolic restriction (Agneessens et al. 2017). Suppose the ratio of added hydrogen to daily anaerobically generated carbon dioxide is less than 8:1. In that case, the concentration of carbon dioxide might be sustained above 11.8%, while the pH increase is limited to 0.17, hence minimising the adverse effects of alkalisation and carbon dioxide consumption.

Exogenous hydrogen addition to the anaerobic bioreactor may increase hydrogen concentration, thereby inhibiting syntrophic biodegradation of volatile fatty acids and

alcohols, resulting in organics accumulation and eventually inhibiting the anaerobic digestion process. As a result, enriching hydrogenotrophic methanogens via long-term acclimatisation or direct bioaugmentation would be a favourable option for rapid hydrogen consumption (Fu et al. 2021). Similarly, conductive substances facilitate the direct transfer of an electron between fermentative bacteria and methanogens (Rotaru et al. 2014) and can establish syntrophic degradation of volatile fatty acids and alcohols, as seen in Eqs. 7 and 8 (Fu et al. 2021). However, additional research is needed to determine whether direct interspecies electron transfer would effectively cancel out hydrogen addition in practice.

One of the most significant challenges of in situ biogas upgradation is increasing system pH. The carbon dioxide dissolved in the liquid part of the anaerobic digestion bioreactor may disassociate into hydrogen ions and bicarbonate (Eq. 5), which might influence the pH through the anaerobic digestion operation (Sarker et al. 2018). Controlling the pH with chemical additives or co-mixing with a low pH feedstock may be an option for overcoming this limitation. Luo and Angelidaki (2013) found that manure co-digestion with acidic whey maintained the pH of the anaerobic digestion system around 7.8 when external hydrogen was introduced for in situ biogas upgrading and that the injected hydrogen was nearly completely utilised for carbon dioxide conversion to methane. Furthermore, ex-situ biogas upgrading by establishing a separate upgrading process from the primary biogas process is a further solution (Voelklein et al. 2019).

Biochar supplements can facilitate methanogenesis during in situ biogas upgrading (Zhang et al. 2020). Biochar amendments may supplement the hydrogenotrophic methanogenic growth with nutrients such as calcium, magnesium, potassium, and phosphorus. For instance, biochar supplementation enhanced *Methanosarcina* and *Methanothermobacter* growth in a thermophilic anaerobic reactor digesting food waste (Zhang et al. 2020). Additionally, biochar has been shown to accelerate electron transfer between syntrophic bacteria and methanogens, thereby increasing biogas production and system stability (Wang et al. 2021a; Wang et al. 2021b). Regular use of biochar may successfully enhance these electrotrophic microbes; additionally, direct interspecies electron transfer may improve volatile fatty acid breakdown (Wang et al. 2021a; Wang et al. 2021b). The fundamental reason for biochar's ability to counteract acidic

and/or alkaline disorder is that functional groups involving carboxylic, amine, and phenolic groups are formed during the pyrolysis process.

Additionally, the metal ions found in biochar, such as potassium and sodium, as well as other "earth" metals, most notably calcium and magnesium, contribute to the buffering value of biochar being maximised (Zhao et al. 2021b). Biochar's attractive buffering capacity is required to compensate for volatile fatty acid accumulation during anaerobic digestion (Wang et al. 2017). Additionally, the role of porous biochar in promoting the development of microbial biofilms and biochar's ability to protect and enrich functional microorganisms attached to biochar under acid stress should be investigated.

Therefore, biochar is proposed as a viable option for hydrogenotrophic upgrading due to the following reasons: (1) biochar can be delivered via an environmentally friendly and cost-effective approach, and (2) biochar physicochemical properties can be tailored to operational conditions (Fagbohungbe et al. 2017a; Chiappero et al. 2020). The full details of the role of biochar in biogas upgrading are discussed in Sect. 2.3.3.

In comparison to in situ biogas upgrading, ex-situ biogas upgrading enforces fewer metabolic constraints and exhibits greater flexibility. Whereby the biogas is upgraded ex-situ in a separate bioreactor without affecting the anaerobic digestion process. Additionally, the biological activities contained within are easier to adjust and control with minimal organic modification, as well as more adaptable to remote control via the power source, allowing for increased control of the integrated system (Angelidaki et al. 2018).

Improving hydrogen mass transfer and utilisation efficiency Another significant constraint on applying in situ and ex-situ biogas upgradation systems is a lack of hydrogen mass transfer. According to Eq. 6, the rate of hydrogen gas to liquid is linearly related to the gas transfer coefficient. Thus, the hydrogen transfer rate can be increased by increasing the L_gc value, which is typically dependent on the operating conditions and configuration of the bioreactor (Bassani et al. 2017; Rusmanis et al. 2019). Different configuration system and operational conditions are summarised as follow:

Continuous stirring tank reactors are the most popular reactor form used in the anaerobic digestion operation. The reactor is equipped with impellers operated by a motor to stir the reactor. Gas production and feedstock retention times can be maximised through mixing speed control. Intense mixing typically requires high energy input and might disintegrate biomass structures and the microorganism's cell, which causes volatile fatty acids build-up in the system (Rusmanis et al. 2019; Wahid and Horn 2021). Thus, the optimal speed of 140–170 rounds per minute has been

suggested to generate a methane rate of 0.9 L/l$_{reactor}$/day (Wahid and Horn 2021). Improved mixing speed increased methane content from 69 to 77%, while gas recirculation at 12.2 millilitres/minute increased methane contents from 77 to 80% (Wahid and Horn 2021). However, this trial does not satisfy the methane quality for the natural gas grids. Luo and Angelidaki (2012) equipped a continuous stirrer reactor to maintain liquid consistency and employed a gas unit for injecting hydrogen and biogas. After increasing the stirring speed from 500 to 800 rounds per minute, the biogas introduction rate raised from 12 to 24 L/l$_{reactor}$/day; in addition, the methane value was about 90%.

Nevertheless, rates of hydrogen gas transfer remained the limiting factor for increased hydrogen utilisation in the system. Furthermore, extreme mixing disrupts synergistic relations between fermenters bacteria and hydrogen-utilising methanogens, thereby hindering fermentation and methanogenesis bioprocesses. To overcome lower hydrogen gas transfer rate, a sequence of new reactor configurations has been implemented and reviewed in some studies (Lai et al. 2021; Wu et al. 2021; Zhao et al. 2021a), including solid-state bioreactor, bubble column reactor, hollow-fibre membrane biofilm reactor, trickling bed reactor, and bioelectrochemical system. This study focused on some configurations that meet our objectives, as seen below.

Bubble column bioreactor. This reactor has supplied with gas diffusers usually made from alumina ceramic membranes or stainless steel for dispersal of hydrogen gas. The transfer rate of hydrogen mass could improve by adjusting the pore size of the diffuser, such as changing the diameter from 2 to 0.4 μm, changing the gas circulation rate, and fixing a ceramic sponge over the hydrogen gas disseminators (Bassani et al. 2017; Bassani et al. 2016). Using *bubble column reactor* for ex-situ upgradation attained methane outcome of 0.25 l$_{methane}$/l$_{hydrogen}$, with a 96% biomethane purity (Bassani et al. 2017). In addition, this configuration could produce more than 98% methane content in the exhaust gas compared to 79% methane content in the continuous stirrer tank system treating the same gas loading rate (Kougias et al. 2017).

The integration of this reactor design with hydrogen nano-bubble (50–200 nm) technology has been suggested by some researchers to improve gas solubility, enhance the contact surface charge between gas and bacteria, and might enhance enzyme activity (nano-carrier) (Fan et al. 2020; Wang et al. 2019; Lyu et al. 2019), but the hydrogen nano-bubble technology is still in early stages, and future research on such integration with hydrogenotrophic methanogenesis is required. Using modified biochar as a dispenser media would be a promising option in this case and would be a future area of study.

Solid-state reactor. The exclusive idea of a *solid-state reactor* is filling solid particles such as granular perlite and vermiculite for trapping methanogens functional microbes.

The features of high particles' specific surface area create adequate contact between gas, methanogens, and liquid, improving the rate of gas transformation (Rittmann et al. 2015). This system could achieve a maximum methane production rate of 6.35 L/l$_{reactor}$/day and a hydrogen utilisation rate of 100% (Alitalo et al. 2015). Using biochar as a solid packing material would be a promising option in this case to support the growth of methanogens and is a promising area of research for the future.

Bioelectrochemical technique for biogas upgrading Biogas upgrading via a bioelectrochemical system has attracted great attention due to various benefits, including waste recovery, low energy input, reduced sludge production, no need for aeration, and the formation of value-added substances (Wang et al. 2022a; Aryal et al. 2022). Carbon dioxide transfer assisted by bioelectrochemical systems can withstand fluctuations in electricity supply, overcoming the instabilities associated with renewable energy (del Pilar Anzola Rojas et al. 2018).

A bioelectrochemical system is typically equipped with a bioanode for oxidation and a biocathode for reduction reactions, which are frequently separated by an ion-transport membrane for the purpose of exchanging ions. The bioanode acts as an electron acceptor, allowing electrotrophic microbes to oxidise organic matter (Zhao et al. 2021a; Aryal et al. 2022). The gathered electrons are transported to the biocathode via an exogenous stimulus, where they are used to convert the targeted byproducts into value-added fuels and chemicals. Typically, organics or wastewater are decomposed at the bioanode compartment, and then the producing electrons are collected at the biocathode, where hydrogen is produced and then used by methanogens to convert carbon dioxide to biomethane (biogas upgrading) (Lai et al. 2021; Zhao et al. 2021a).

Electromethanogenesis can occur via several pathways: First, through the interspecies electron exchange, where the generated H$^+$ at the biocathode is catalysed by extracellular enzymes to produce hydrogen, which is further captured by hydrogenotrophic methanogens for converting carbon dioxide into methane. Second, the electron can be directly transferred and used by methanogens to reduce carbon dioxide into methane by physical contact on biocathode without the intermediate hydrogen production (Fu et al. 2021; Hagos et al. 2018). Finally, the *Homoacetogens* can convert the carbon dioxide and hydrogen intermediates into acetates, which then are utilised by the acetoclastic methanogens to generate methane. Overall, interspecies electron exchange is more dominant in electromethanogenesis, which enables the bioelectrochemical system to overcome some extent of the gas-to-liquid transfer limitations in external hydrogen injection (Zhao et al. 2021a).

Usually, the external hydrogen addition in an in situ upgrading system leads to the accumulation of about 6 g/litre of acetate (Tartakovsky et al. 2021). This issue was not noticed in the bioelectrochemical system, proposed the bioelectrochemical system to be promising and of superior performance (Tartakovsky et al. 2021). Moreover, electrochemical, or microbial hydrogen sulphide oxidation can remove the hydrogen sulphide produced from the raw substrates with sulphate or sulphur (Lai et al. 2021; Ni et al. 2019).

The bioelectrode in the bioelectrochemical system showed higher biocathode-related biomass, which was proved from protein evaluation of biofilms produced at the biocathode. The hydrogenotrophic methanogens were the major leading species in the microbial population (Bo et al. 2014). Hence, the in situ hydrogen production by using the bioelectrochemical system could boost hydrogenotrophic methanogens' activity, thus altering the microbial dominance (Cerrillo et al. 2021; Gao et al. 2021).

At the bioelectrochemical system, the relative abundance of hydrogenotrophic methanogens such as *Methanobacteriales* and *Methanomicrobiales* was improved up to 17.2 folds (Gajaraj et al. 2017). In addition, *Methanobacterium* was the most abundant microbes at both batch and continuous modes (Aryal et al. 2022). Hence, selective improvement for hydrogenotrophic methanogens caused by hydrogen generation in the bioelectrochemical system can be assumed. In addition to the prevailing of hydrogenotrophic methanogens, acetoclastic is also present in the bioelectrochemical system to utilise acetate into methane and carbon dioxide; also, the presence of carbon dioxide and hydrogen could activate the *Homoacetogens* to produce acetate (Aryal et al. 2022). The factors affecting the efficiency of the carbon dioxide conversion into methane using a bioelectrochemical system are listed in Table 11.

In batch and continuous operation, reactors with an H-shape were frequently used for biogas upgrading (Fig. 6). To achieve that configuration, the reactor with a membrane divided the bioanode from the biocathode was commonly used in a double-chambers reactor for in situ and ex-situ biogas upgradation (Wu et al. 2021; Aryal et al. 2022). In the in situ bioelectrochemical-supported biogas upgrading mode, bioelectrodes were incorporated into the anaerobic digesters to stimulate hydrogen or electron generation for carbon dioxide conversion (Fig. 6a). Simultaneous degradation of organic particulate into methane may occur, while the generated oxygen may de-sulphurise the biogas and aid in organic hydrolysis (Lai et al. 2021).

In ex-situ bioelectrochemical-assisted biogas upgradation, the outcoming biogas from an anaerobic fermenter is delivered into the bioelectrochemical unit, where carbon dioxide from raw biogas is converted to methane either directly by accepting the electrons from the electrodes or indirectly via hydrogen integration pathway (Fig. 6).

Table 11 Role of the bioelectrochemical system in biogas upgrading

Biocathode material	System mode/upgrade method	Bioreactor configuration	Inoculum and dominant microorganisms	Current density	Upgrading/improvement	Biocathode efficiency (%)	References
Stannic oxide nanoparticles	Batch/ex-situ	Anaerobic membrane bioreactor plus microbial electrolysis cell separated by the membrane	Wastewater treatment plant sludge culture (*Methanobacterium*, *Methanosaeta*, and *Methanobrevibacter*)	7.2×10^4 milliamperes per square metre	90% methane	85	Gao et al. (2021)
Carbon cloth	Ex-situ	Microbial electrolysis cell plus hydrogen injection to anaerobic digestion	Methanobacteriales Methanosarcinales Methanomicrobiales	0.4 amperes per square metre	Methane yields 500 millilitres/gram chemical oxygen demand	Not given	Yanuka-Golub et al. (2019)
Carbon paper	Batch/ex-situ	Two chambers	*Methanococcus maripaludis*	Not given	98.3% methane	85.2	Fu et al. (2020)
Graphite plate	Batch/in situ	Two chambers plus a membrane	Wastewater sludge cultures (*Methanobacterium* and *Methanothrix*)	0.022 milliamperes per square metre	97% methane	68.1	Liu et al. (2021)
Carbon paper	Batch and continuous/ in situ	Two chambers plus proton exchange membranes	*Methanococcus maripaludis* S2	Not given	98.3% methane	85.2	Fu et al. (2020)
Graphite granules	Batch/in situ	Microbial electrolysis cell based on hydrophobic membranes	Electromethanogenic culture	$12.1 \pm 4.8 \times 10^4$ milliamperes per cubic metre	73 ± 8 L methane/day (1.5 times enhanced)	23	Cerrillo et al. (2021)
Carbon brush	Batch/in situ	Microbial electrolysis cell divided by a membrane	Municipal wastewater treatment plant culture (*Methanobacterium* and *Methanosarcina*)	0.407 milliamperes per square metre	91.2% methane	18.8	Liu et al. (2020a)
Carbon felt	Batch/ex-situ	Two chambers plus microbial electrolysis cell membrane divided by nylon cloth	Anaerobic sludge culture	250 milliamperes	90% methane	Not given	Tartakovsky et al. (2021)
Graphite granular	Ex-situ	Three separations membrane	Thermophilic anaerobic culture	145 ± 4 milliamperes 129 ± 3 milliamperes	68 ± 6 mmol 71 ± 14 mmol carbon dioxide removal	69 ± 4 95 ± 4	Zeppilli et al. (2019)
Graphite plate	Continuous experiment/in situ	Anaerobic digestion plus microbial electrosynthesis with two chambers	Wastewater treatment plant sludge culture (*Methanobacterium* and *Methanothrix*)	Not given	More than 90% methane	97.60	Liu et al. (2019)

Table 11 (continued)

Biocathode material	System mode/upgrade method	Bioreactor configuration	Inoculum and dominant microorganisms	Current density	Upgrading/improvement	Biocathode efficiency (%)	References
Carbon fibre	Batch/in situ	Single chamber	Wastewater treatment plant sludge culture	15.54 milliamperes	0.34-L methane/gram chemical oxygen demand	Not given	Lee et al. (2019)
Graphite loaded with iron, copper and nickel	Batch/in situ	Anaerobic digestion plus microbial electrolysis in a single chamber	From food waste treatment plant originated culture (*Methanobacterium Methanosarcina*)	Not given	1.7-time faster than anaerobic digestion	Not given	Park et al., 2018
Stainless steel	Batch/in situ	Multi-chambers microbial electrolysis cell	Wastewater treatment plant sludge culture	1490 milliamperes per square metre	99–100% methane	Not given	Kokkoli et al., 2018

Various in situ/ex-situ biogas upgradation systems incorporated with bioelectrochemical systems are used. The biocathode integrated with single, two, or triple chambers showed methane enhancement from 90 to 100%. Carbon-based fabrics are the best applicants for bioanode and biocathode manufacture. The membrane design increases the efficiency of electrons utilisation for methane generation by a bioelectrochemical system. Multi-chamber reactor configuration produces higher methane at the biocathode chamber than the single chamber configuration. Almost the experiments are conducted in batch mode; however, large-scale application is missed

An innovative two or three-chambers bioelectrochemical system was established to upgrade biogas and treat organic wastes (Fig. 6b, c). In the three-chambers design, an anion exchange membrane and a cation exchange membrane were installed to separate the system into two biocathode chambers (on two sides) and one bioanode chamber (in the middle), as illustrated in (Fig. 6c). Biogas is flown into the cation exchange membrane (biocathode) chamber, while feedstocks containing organics are fed into the anode chamber. Organics were disintegrated in the bioanode compartment, while carbon dioxide in the biogas stream was eliminated in both biocathode chambers through carbon dioxide adsorption and methanogenesis (Zeppilli et al. 2019).

The methanogenesis in the biocathode might also migrate ammonium ions from the bioanode for supporting electroneutrality, thus facilitating the ammonium recovery in this compartment (Fig. 6c). Lately, Fu et al. (2020) established a bioelectrochemical system supported by a proton exchange membrane and spiked ferrous ion into the bioanode chamber, aiming to upgrade biogas and recover the sulphur. When biogas was introduced into the bioelectrochemical system, sulphide form was oxidised to sulphur via a ferrous ion-facilitated redox interaction in the bioanode chamber (Fu et al. 2020). Meanwhile, hydrogen was produced by integrating electrons from the biocathode and protons that transferred from the bioanode throughout sulphide oxidation. The hydrogen produced was then utilised to drive carbon dioxide reduction to methane, assisted by the hydrogenotrophic methanogens action in the biocathode chamber.

Some reports showed the superiority of the in situ bioelectrochemical system over the ex-situ for biogas upgradation. Xu et al. (2014) found that the carbon dioxide utilisation rate was higher in in situ than in the ex-situ biogas upgradation process. The current density demonstrated that the quantity of the charge utilised per electrode unit for the utilisation of carbon dioxide was 0.4 A per square metre in ex-situ, compared to 1 A per square metre for in *situ*. The authors attributed carbon dioxide gas–liquid mass transfer limitations to lower current density in the ex-situ system. The in situ systems did not expose to those limitations because carbon dioxide is released by organic particulate biodegradation occurred in the bioelectrode chamber. Equally, the in situ single compartment bioreactor has displayed better performance in current density and biogas upgrading due to more affordable biomass and nutrients established on the electrode upgrading's surface (Aryal et al. 2022; Lee et al. 2019).

In in situ bioelectrochemical mode, multiple anaerobic fermentations are contained in carbon dioxide reduction; in addition, multiple biomass exists. However, in the ex-situ systems, only carbon dioxide decline is targeted, and electricity input is the only energy source. Therefore, the operational mode is critical for selecting the reactor configuration. Although the in situ bioelectrochemical system

Fig. 6 Various bio-electrochemical configuration approaches. **a** Single chamber design; **b** Two-chamber configuration; and **c** represents three-chamber configurations. An ion exchange membrane is used to divide the bioanode from the biocathode. The carbon dioxide from raw biogas is converted to methane either directly by accepting the electron from the electrode or indirectly through the hydrogen integration pathway. In the three-chambers design, two exchange membranes are installed to separate the system into two biocathode chambers and one bioanode chamber. Biogas is flown into the biocathode chambers, while feedstocks containing organics are fed into the bioanode chamber. The methanogenesis process in the biocathode may migrate ammonium ions from the bioanode to improve ammonium recovery in this chamber
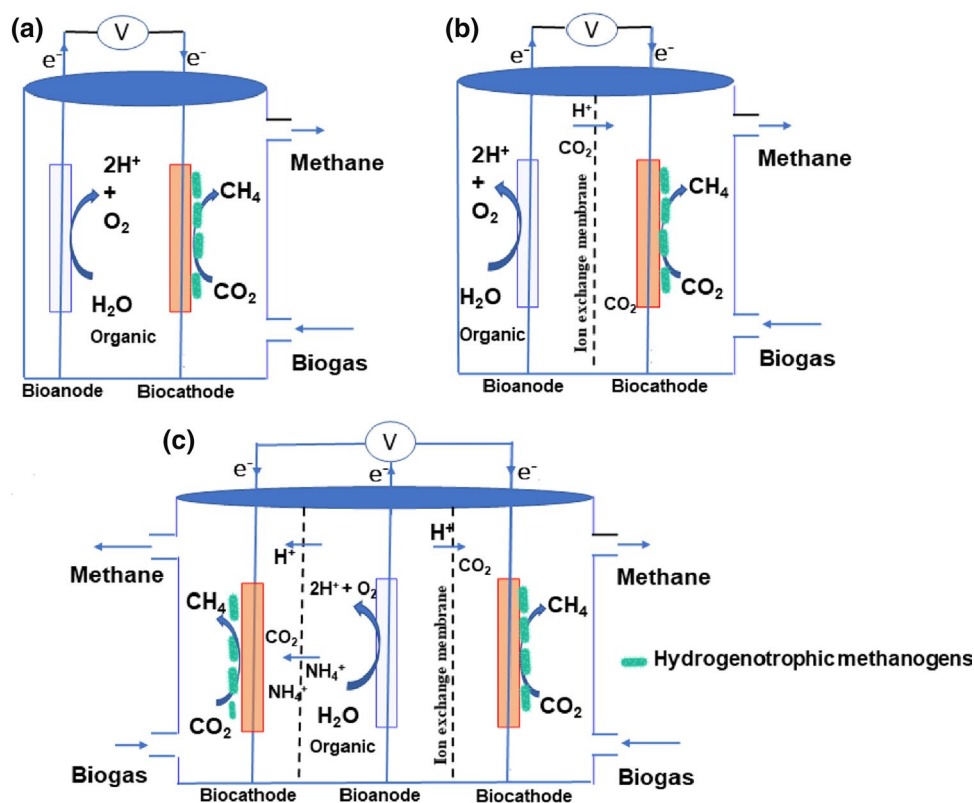
indicated several merits for scaling up the bioelectrochemical system to upgrade biogas from anaerobic digestion over ex-situ, the small conformation size of an in situ bioelectrochemical system is the bottleneck because the bigger-sized reactors probable bring lower current density and reducing hydrogen generation rate (Wu et al. 2021). Furthermore, basic studies such as examining the electrode-microbes interface, the impact of membranes, and electron transfer way need to be performed in in situ and ex-situ in the future. Other considerations, such as carbon dioxide utilisation rate, pH, the effect of membrane on biogas upgrading, and methane purity, should be deemed before concluding the dominance of in situ over ex-situ mode.

In summary, the realisation of a bioelectrochemical system for biogas upgrading is dependent on the reactor configuration and electrode materials. Carbon-based fabrics are the best candidates for manufacturing bioanodes and biocathodes. The membrane design improves the efficiency with which electrons are used to generate methane in a bioelectrochemical system. Due to their optimistic effects on combined biofilm growth and hydrogen generation, a new electrode configuration is required to achieve a higher p potential for carbon dioxide conversion from biogas.

Summary

In summary, several methods for biogas upgrading have been developed, including physical, chemical, and biological approaches. The physical upgrading technique is based on removing carbon dioxide from raw biogas via water/organic scrubbing, pressure swing adsorption, membrane separation, or cryogenic separation. At atmospheric pressure and room temperature, the biological biogas upgrading technique utilises chemoautotrophic reactions in which microorganisms act as catalysts to facilitate the conversion of carbon dioxide to methane or other valuable byproducts. Physical and chemical upgrading techniques are commercially viable in the biogas industry due to their increased efficiency, selectivity, and biomethane content in the upgraded biogas. Nonetheless, these techniques have several disadvantages, including high energy and investment requirements, hazardous chemicals, and the requirement for external energy.

Additionally, the carbon dioxide emitted by physical biogas upgrading technologies is emitted directly into the environment, which is not only a waste of a carbon source but also contributes to global warming. Alternatively, biological upgrading technology has garnered considerable interest as a result of the exceptional benefits associated with reduced energy and carbon footprints. The critical advantage of the biological biogas upgrading approach is that carbon dioxide is captured and repurposed for new products via

carbon dioxide reaction with hydrogen to form biomethane. Hydrogen can be obtained through water electrolysis, and the required electricity can be generated using renewable wind, solar, or hydropower resources. This strategy proposes a concept of power-to-gas conversion that improves energy storage and fosters the development of an environmentally friendly, sustainable, and circular economy.

While the benefits of hydrogen-assisted chemoautotrophic biogas upgrading have been discussed previously, the low hydrogen mass transfer rate between the gaseous and liquid phases is a critical technical constraint on biological upgrading systems. The release of unused hydrogen due to this method would increase the risk of an explosion. Additionally, increasing the pH value above 8 due to methanogenic carbon dioxide oxidation can slow or disrupt microbial bioprocesses, most notably syntrophic correlations between acidogenic and methanogenic microbes. As such, several approaches are recommended to increase the efficiency of hydrogen-assisted upgrading and avoid the issues:

- Hybrid hydrogen-assisted biological upgrading technology combines the anaerobic conversion of carbon dioxide to biomethane with the biomethanation of residual carbon dioxide in a separate unit. This approach has the potential to reduce the low mass transfer of unused hydrogen. Because the hybrid approach is a theoretical concept with limited experimental data, a hybrid approach is recommended for future research.
- Integrating biochar-based additives with hydrogen-assisted biogas upgrading via biological technologies is an extremely promising area for future research.
- Biochar as packing material for solid-state bioreactors and the bioelectrochemical system would allow better integration. Such integration strategies can make use of carbon dioxide and reveal several benefits: (1) wastes can be efficiently converted to energy resources; (2) generated gas can be delivered and stored in gas grid pipelines; (3) biomethane can be used directly for renewable energy and transportation fuel; (4) utilising existing energy infrastructure would be an economically viable option.

Solid digestate as a carbon sequestration tool

Sanitary landfilling and incineration are the two primary waste treatment technologies currently in use; however, incineration produces ash residues and toxic emissions, whereas landfilling produces greenhouse gases (Logan and Visvanathan 2019); As a result, anaerobic digestion is a more environmentally friendly method of waste management (Gao et al. 2017; Liikanen et al. 2018; Zhou et al. 2018). Where anaerobic digestion has the potential to stabilise organic

wastes, reduce their carbon footprint in the environment, and serve as a viable alternative to waste landfill pollution (Khan et al. 2021; Hunter et al. 2021), and can also generate electricity, biomethane, and heat, allowing for a more rapid global deployment of anaerobic digestion operations (Jain et al. 2019; Bioenergy 2022). Thus, anaerobic digestion can contribute to decarbonisation and defossilisation by capturing biomethane and substituting fossil fuels (WBA 2021).

Digestate effluent, anaerobic digestion of nutrient-rich byproducts, is a mixture of undigested substrates, metabolites, inert organics, and microbial biomass (Cavali et al. 2022; Guilayn et al. 2022). Using nutrient-dense digestate (i.e. phosphorus, potassium, nitrogen, and other micronutrients) from agricultural waste, such as energy crops, livestock manure, agricultural residues and straw, and others, is widely accepted as a soil biofertiliser to promote crop growth and land health. However, the massive quantities of digestate produced by anaerobic digestion facilities and proper management have raised concerns about valorising this byproduct, whereas without proper management policies, the digestate of anaerobic digestion contributes not only to nutrient pollution, such as eutrophication, harmful algal blooms, and hypoxia (Lamolinara et al. 2022) but may also result in a variety of environmental risks, such as pathogen spread and heavy metal pollution (Logan and Visvanathan 2019; Peng and Pivato 2019) and substantial greenhouse gas emissions (Peng et al. 2020a, 2020b). As a result, managing the anaerobic digestion effluent digestate in a way that ensures an environmental and circular economy is currently a bottleneck for the sustainability of biogas plants (Peng et al. 2020b).

Current post-treatment technologies for digestate

There are numerous methods for digestate processing, which are generally determined by the digestate's physicochemical characteristics and intended use (Ma et al. 2018; Zubair et al. 2020). The currently available digestate processing options can be classified by type into physical (e.g. settling, flotation, screening), biological (i.e. bioremediation), or chemical treatment (e.g. oxidation processes); secondly, by the portion of the digestate applied to a liquid or solid separation; and thirdly to partial or a complete upgrading (Lamolinara et al. 2022; Herbes et al. 2020). A partial end-use policy seeks to minimise volume, whereas a fully processed digestate policy seeks to refine the digestate to solids or fibres, pure water, and mineral concentrates (Logan and Visvanathan 2019). Solids separation from the liquid fraction is the first stage in digestate processing, as assumed in Table 12. However, many of these technologies are prohibitively expensive due to low material efficiency, high energy requirements, and initial development stages (Herbes et al. 2020).

Table 12 Digestate treatment technologies (Herbes et al. 2020)

Technology used	Raw digestate	Solid fraction	Liquid fraction
Physical	Ultrasound Solar dryer Belt and drum dryers Screw press	Drum dryer Belt dryer	Ammonia stripping (physicochemical process) Reverse osmosis Vacuum evaporation Ultra-filtration Micro filtration
Biological	Algae Composting	Composting	Algae
Chemical	Flocculation	Not applicable	Flocculation Struvite precipitation

The digestate treatment methods currently available are summarised. These can be classified into three types of treatment: physical, chemical, and biological. As a qualification for further treatment, the digestate may be separated into solid and liquid components in order to produce a consistent biofertiliser (liquid or solid) that improves the digestate marketability and quality. The digestate treatment approaches are designed to remove organic matter and nutrients from the waste stream while allowing for the production of secure effluent. Additionally, the latter approach provides comprehensive treatment, including a solid biofertiliser, purified water, and concentrated mineral nutrients. Both treatment and conditioning are required to produce a viable digestate post-treatment

Numerous commercially available solid–liquid separation methods include centrifuges, decanter screw press separators, bow sieves, sieve drums, and sieve belt presses (Logan and Visvanathan 2019; Guilayn et al. 2019). The screw press separator and decanter centrifuge have gained popularity among farmers who export excess nutrients. Additionally, decanter centrifuges are frequently used in municipal waste treatment plants, whereas screw press separators are primarily used for digestate-rich fibres.

In general, the first phase of any digestate processing system is solid–liquid separation. This separation typically yields a solid portion, referred to as a press cake, with a dry matter content of 20–30% and a liquid portion with a dry matter content of approximately 3%. The liquid fraction is primarily composed of mineral nitrogen and potassium, while the solid fraction is predominantly composed of organic nitrogen and phosphorus (Herbes et al. 2020). Due to the high humus and fertiliser value of the phosphorus-rich solid fraction, solid digestate is frequently transported to remote regions. Solid digestate fraction can be dried, pelletised, composted for use as soil fertiliser, or used industrially or incinerated for energy recovery (Logan and Visvanathan 2019). The low organic content and high-water content have limited market potential for the liquid portion of digestate. In some cases, both fractions are used directly as fertiliser.

Drum dryers, belt dryers, and solar dryers are currently used in the market. Generally, a belt dryer is used to dry the press cake and is typically combined with ammonia scrubbing of the output air (to prevent ammonia emissions) (Herbes et al. 2020; Awiszus et al. 2018). Solar drying is accomplished by transporting or pumping the solid fraction from the separation process or even the raw digestate into a greenhouse and allowing the water to evaporate through the action of solar radiation. The substrates are frequently mixed, and floating fans exhaust the greenhouse's water-saturated air (Maurer and Müller 2019). This method can achieve a dry matter content of approximately 65% in biogas plants. However, because a large volume of air must diffuse through the greenhouse, air scrubbing is, in most cases, technically impractical. Additionally, the subsequent ammonia releases have a detrimental effect on the environment and a significant decline in fertiliser value (Herbes et al. 2020).

Overall, digestate processing techniques are expensive and energy-intensive. Membrane-based treatment is also prohibitively expensive; additionally, all drying and evaporation processes rely on heat (Herbes et al. 2020). Chemical treatments of digestate create complications regarding the reuse and recovery of additives or chemical reagents used. The producing digestate must be converted into a sustainable fertiliser while adhering to the concepts of reducing, reusing, and recycling with the least amount of environmental impact possible.

Limitations of digestate land spreading

The most prevalent valorisation pathway for anaerobic digestate is digestate use in agronomic soils. If digestate is likely managed properly, digestate has the potential to replace inorganic chemical fertilisers, thereby alleviating subsequent environmental concerns (Panuccio et al. 2019; Verdi et al. 2019). However, direct land spreading is the most widely used strategy for valorising anaerobic digestate management.

Common limitations for the direct land spreading of digestate can be summarised as follow:

1. Digestate frequently contains partially degraded biodegradable organic matter as well as complex organic pollutants such as fungicides, herbicides, hormones, industrial wastes, excessive salt concentrations, and pathogens (Lamolinara et al. 2022).

2. Due to the presence of biological contaminants in digestate, disease and pathogens can be transmitted in various ways between humans, animals, and the environment. As a result, strict control of feedstock and digestate is required. Animal byproducts used as anaerobic digestion feedstock require special consideration in terms of their safe use as soil conditioners and fertilisers (Logan and Visvanathan 2019).

3. One significant obstacle to digestate production is that the amount of effluent digestate produced may exceed the capacity of the region's existing arable soils to fertilise (Logan and Visvanathan 2019; Guilayn et al. 2022; Nkoa 2014; Vaneeckhaute et al. 2013), the factor that contributes to an increase in the cost of transportation. Dahlin et al. (2015) demonstrated that the digestate transfer distance had been increased by a factor of two over the last few years, reaching 150 km. Additionally, the number of centralised and large biogas plants has increased as their environmental and economic feasibility has increased. Nonetheless, most of these facilities exceed the local demand for nutrient spreading. The timing and amount of digestate applied are largely determined by the characteristics of the soil and the plants being cultivated. Additionally, according to crop growth season, adequate digestate storage facilities must be recognised to manage digestate production. The requirement for digestate storage is greatest when crop cultivation is limited in season and is negligible when crop growth is adequate throughout the year (Logan and Visvanathan 2019). The massive amounts of digestate are due to digestate higher water content, making storage difficult and expensive transportation (Herbes et al. 2020; Silkina et al. 2017). As a result, any advancements in solids-liquids separation would be beneficial. Drying the digestate would increase productivity and viability by reducing digestate volume and valorising digestate nutrients. For instance, food waste liquid digestate represented 79% of the feedstock's mass, while after dewatering procedures, the fertiliser product represents only 16% of the biomass's initial mass (Tampio et al. 2016).

4. Digestate may contain heavy metals, such as lead, cadmium, nickel, chromium, mercury, cupper, and zinc, along with organic pollutants, such as polychlorinated biphenyls, accidental pollutants from the industry like dioxins and furans, incomplete combustion products like polycyclic aromatic hydrocarbons, plasticisers (phthalates), personal care products, medicines, pesticides, antibiotics residues, emerging contaminants, among others (Lamolinara et al. 2022). These materials may be toxic to humans, livestock, and ecosystems, and disposing of them is challenging.

5. Digestate must be properly managed, processed, and stored in order to reduce and eliminate pollution emissions such as nitrous oxide, ammonia, methane, and odour (Zilio et al. 2020).

6. The economic value of digestate is critical. Czekała et al. (2020) reported that the daily revenue from a biogas plant with a capacity of 1 megawatt is approximately €1414, and the digestate profit is approximately €334.4. Thus, proper digestate management throughout the year can be a source of revenue that contributes to the biogas plant's profitability. In contrast, the retail price of digestate in Europe is several times lower than the cost of production (€5–30/tonne), which is attributed to the high digestate hydration problem; however, the digestate cost may increase significantly to €250/tonne if digestate is sold in dry pelletised form in smaller containers. As a result, efforts to concentrate the digestate would improve a biogas plant's financial balance (Czekała et al. 2020).

7. Although digestate's greenhouse gas emission potential is approximately 75% lower than that of municipal solid waste's organic fraction (568 g carbon dioxide equivalent/kilogramme waste), digestate still emits greenhouse gases (139 g carbon dioxide equivalent/kilogramme waste) (Logan and Visvanathan 2019). Thus, in order to optimise the anaerobic digestion process from a greenhouse gas emission perspective, digestate emissions must be considered.

8. According to Guilayn et al. (2022); Guilayn et al. (2020), numerous technical and legal bottlenecks exist, particularly for non-agricultural digestates. For instance, the European Union's fertilisers regulation (CE 2019/1009) bans the use of certain municipal feedstocks such as sewage sludge, sewage sludge organic fraction, and other mixed wastes. Such legislation is necessary to enshrine sound waste disposal. Additional regulations addressing renewable energy demand, global warming, organic waste landfill tax, high fossil fuel prices, organic fertiliser demand, and environmental pollution may have an effect on digestate management policy. In some countries, regulations promote decentralised anaerobic digestion by restricting land use to digestate generated on-farm (Logan and Visvanathan 2019).

9. Another disadvantage of anaerobic digestion is the diminished nutritional value of the digestate residual effect following the separation of the solid and liquid fractions (Masebinu et al. 2019). Likewise, the instability of anaerobic digestates contradicts the environ-

mental sustainability theory due to methane emission into the environment (Fagbohungbe et al. 2017b).

10. Direct spreading of digestate is frequently insufficient to overcome digestate challenges, particularly in the case of non-agricultural digesters or large-scale digesters. As a result, novel approaches to digestate valorisation are critical (Guilayn et al. 2022; Guilayn et al. 2020).

11. Apart from technical constraints, the varied quality of digestates, public acceptance, and difficulties in establishing new markets all pose significant barriers to valorising anaerobic digestion into value-added end products (Guilayn et al. 2020).

As an alternative, this review focuses on the use of dried solid digestate to produce biochar as a carbon sequestration step and on ensuring solid digestate recycling, reusing, and size reduction in the biogas and agriculture fields.

Digestate-driven biochar

Anaerobic digestates are massive organic byproducts of anaerobic digestion. Prior to final disposal, digestate waste streams must be treated to reduce volume, hazardous biowaste such as pathogens, and offensive odours. Traditional methods of removing anaerobic digestate, such as direct application to agriculture or landfilling, may be limited due to the possibility of pollutants being transported to the land and the scarcity of landfill sites, respectively. As a result, managing massive amounts of digestate produced by anaerobic digestion has become a critical issue in conducting a comprehensive assessment of biogas facilities in terms of digestate processing and treatment.

One option for upgrading digestates is their thermal conversion to biochar, which is currently being developed. Numerous benefits are anticipated from digestate conversion into biochar concept, including organic waste stabilisation, nutrient and carbon sequestration and reuse, conserving natural resources, being energy-positive, cost-effective, adaptable to a variety of substrates, ensuring controlled product quality, generating value-added byproducts from high moisture feedstocks, increasing social acceptance, and establishing a new market economy.

Biochar is a carbon-rich solidified char obtained from thermal methods such as gasification or pyrolysis of feedstock in a low or oxygen-free environment (Sakhiya et al. 2020) or hydrothermal carbonisation (named hydrochar). Biochar is distinguished by biochar physical, biological, and chemical properties, including a large specific surface area, an aromatised carbon matrix, a high porosity, a high mineral content, and abundant surface functional groups (Panahi et al. 2020). Hydrothermal carbonisation and pyrolysis/gasification can convert digestate to hydrochar or biochar. Biochars and hydrochars are discussed in this section for their potential applications in four major areas: carbon sink for long-term stability, carbon sequestration, biogas production or upgrading, and pathway integration with anaerobic digestion processes (recycling pathway). The stability mechanisms underlying biochar production are discussed in detail,

Table 13 Operational conditions and different yields from thermal conversion methods (Sakhiya et al. 2020; Zhang et al. 2019; Kambo and Dutta 2015; Belcher 2013; Sohi et al. 2010; Wongrod et al. 2022; Kung et al. 2022)

Thermal process	Operation conditions	Syngas (%)	Bio-oil (%)	Biochar/hydrochar (%)
Fast pyrolysis	Temperature: 400–900 °C Heating rate: 100–1000 °C/s Residence time: second to minute	About 13	About 75	About 12
Slow pyrolysis	Temperature: 250–700 °C Heating rate: 5–30 °C/min Residence time: 15 min–2 h	About 35	About 30	About 35
Intermediate pyrolysis	Temperature: 300–450 °C Heating rate: 3–10 °C/min Residence time: 10–20 s	20–30	35–50	25–40
Gasification	Temperature: 800–950 °C Heating rate: 50–100 °C/s Residence time: 10–20 s	About 85	About 5	About 10
Hydrothermal carbonisation	Temperature: 180–260 °C Heating rate: 5–10 °C/min Residence time: 15 min–2 h	About 5	About 25	About 70

Numerous thermochemical processes can be used to treat biomass and generate various products and byproducts. Fast pyrolysis produces biochar, gas, and oil with % yields of 12, 13, and 75%, respectively, while slow pyrolysis produces 35, 35, and 30%, respectively. Gasification can generate up to 85, 5, and 10% of syngas, bio-oil, and biochar, respectively. Hydrothermal carbonisation produces approximately 70% biochar

as are the major parameters affecting the biochars' carbon sequestration efficacy and potential environmental impact.

Biochar and hydrochar production technologies

Currently, two major techniques for producing biochar are hydrothermal carbonisation and pyrolytic treatment, as seen in Table 13. The pyrolytic carbonisation method operates at a temperature range of 300–900 °C; however, the hydrothermal carbonisation method operates at a temperature range of 180–260 °C. Pyrolysis can be classified into slow pyrolysis, which uses a low heating rate and a long residence time, and fast pyrolysis, which uses a high heating rate and a short residence time. Slow thermal pyrolysis is typically carried out at temperatures between 400 and 600 °C to maximise biochar yield while also producing a small amount of concentrated bio-oil and syngas containing methane, hydrogen, carbon dioxide/monoxide, and hydrocarbons. Additionally, slow thermal pyrolysis is achieved at atmospheric pressure with a long residence time of more than 1 h and heating rates ranging from 5 to 30 °C/min (Zhang et al. 2019; Al Arni 2018; Kambo and Dutta 2015). Slow pyrolysis produces a high yield of biochar, approximately 35%, in addition to syngas and bio-oil yields of 35% and 30%, respectively (Zhang et al. 2019). There is also intermediate pyrolysis, which has a shorter residence time than slow pyrolysis but also a slightly higher heating rate.

Alternatively, fast pyrolysis produced bio-oil as the primary product (75%) and syngas and biochar as by-products (10–15% each). Although bio-oil has a high viscosity and a low heating value, bio-oil can be improved for use in various fuels and valuable chemical applications (Akhtar et al. 2018). The fast thermal pyrolysis technique operates between 400 and 700 °C with an extremely high heating rate of approximately 1000 °C per second and a very short residence time of less than a minute (Zhang et al. 2019; Mohan et al. 2014). In general, fast pyrolysis produces less biochar and has higher operating costs; thus, slow or intermediate pyrolysis is more advantageous for producing biochar (Pituello et al. 2015).

The gasification process is primarily used for gas generation (Zhang et al. 2019; Novotny et al. 2015). The fuel gas produced comprises carbon monoxide and hydrogen that may be used as renewable energy resources for power supply and core engine industries (Zhang et al. 2019; Novotny et al. 2015). However, the extremely high working temperatures (up to 950 °C) resulted in a relatively small amount of biochar, making biochar less attractive from a biochar standpoint.

Combining thermal treatments and anaerobic digestion is a viable option not only to increase anaerobic digestion energy productivity but also to increase the digestate value cascade. Table 14 shows the potential conversion of digestate into biochars and hydrochars.

In general, the biochar yields are inversely proportional to temperature increase (Hu et al. 2022; Liu et al. 2020b; Belete et al. 2021; Cao et al. 2019; Peng et al. 2022). Yields decreased gradually when increasing the reaction time (Xie et al. 2022). The increase in solid loading rate increased the biochar contents. Hung et al. (2017) stated that the low higher heating value of pyrolytic biochar derived from swine manure digestate was attributed to biochar's ash content, which limited pyrolytic biochar use as solid biofuel. Nevertheless, the obtained biochar might be used for soil amendment due to the abundance of alkaline surfaces that can help balance acidic soil pH. Additionally, if the digestate-derived biochar originated from a nutrient-dense waste, such as nitrifying-enhanced sludge, digestate-derived biochar application to the soil would be more beneficial for fertilisation schemes (Sepehri and Sarrafzadeh 2018).

Alternatively, biochar produced through the pyrolysis of solid food waste exhibited a macropore and mesopore arrangement with relatively large specific surface areas (Liu et al. 2020b), which may be appropriate as sportive means for environmental pollutants remedy. The functional groups formed on the biochar's surface are able to adsorb contaminants from waterbodies and the soil environment (Song et al. 2021), including antibiotics, heavy metals, dye and herbicide, along with ammonia and phosphates (Kumar et al. 2020b). Additionally, modified biochar can catalyse various environmental purposes (Wan et al. 2020; Wang et al. 2020; Kumar et al. 2020c).

Another available option is to use the gasification for digestate charring (Zhang et al. 2021; Chen et al. 2017), yet this process produces smaller quantities of biochar as a byproduct, while the syngas is the main product. Syngas can be used to generate electricity through boilers or generators, as well as for a variety of chemical applications as a building block for higher value-added products (Akhtar et al. 2018; Yao et al. 2017). Rather than that, biochars produced by gasification of food waste digestate exhibited excellent surface functionalisation and high phosphorus, potassium, and nitrogen values, indicating that they are well suited for use in soils or as an adsorbent for environmental applications (Zhang et al. 2021). Biochars may achieve carbon sequestration when used in soils due to high fixed carbon contents attained after gasification (Cavali et al. 2022; Chen et al. 2017). Additionally, biochar enhances the soil's nutrient and water retention capacity, thereby reducing greenhouse gas emissions and fertiliser use (Song et al. 2021).

Drying the digestate (65–80 °C) prior to pyrolysis or gasification is required to reduce the moisture content of the digestate to less than 10% (Wongrod et al. 2022). Before pyrolysis, the biomass/digestate is dried, and gasification is an energy-intensive process, which is considered an

Table 14 Potential production of biochar and hydrochar from anaerobic digestate

Digestate supply	Thermal operation and temperature	Heating rate and residence time	Outcome (%)	Char properties	References
Agricultural residue Sewage sludge Municipal solid waste Vegetable, garden, and fruit waste	Hydrothermal carbonisation at 150, 200, and 250 °C	1 h	47.2–82.7 65.6–87.1 82–90.6 71.1–89.0	Yield reduced with increased temperature and decreased solid loading rate Higher carbon and lower oxygen contents than raw digestate (maximum fixed carbon is 21.6 for agricultural residue) Higher ash content with solid loading rate and pyrolysis temperature increase	Parmar and Ross (2019)
food waste and corn husk	Pyrolysis at 900 °C of different mixing ratios	1 h; 30 °C/min	22.75–52.75	Yield is reduced with reduced food waste contents in the mixture	Chen et al. (2022b)
Agricultural waste, manure from animals and poultry, and sludge	Pyrolysis at 400 to 700 °C	10, 20, 30, and 40° C/min	59.26–75.95	Biochar yields decreased with the increased temperature. Pyrolysis gas yield rose	Hu et al. (2022)
Sugar beet bagasse, maize silage, manure, and fruit pomace	Pyrolysis at 400–800 °C	25 °C/min; 5 h	29.42–39.63	Yield, nitrogen, hydrogen, oxygen, and polarity of biochars are inversely correlated with pyrolysis temperature increase	Stefaniuk and Oleszczuk (2015)
Maize silage, distiller grains, potato pulp, manure, and sugar beet bagasse			27.16–30.57	Electrical conductivity, pH, ash, macro/micronutrients, and biochar aromaticity are inversely correlated with temperature increases	
Maize silage, fruit pomace, manure, and sugar beet bagasse			37–45.27	Using the digestate solid fraction increased the carbon (2.57%) and nitrogen (67.55%) contents Surface areas obtained ranged from 0.1 to13.4 square metres/gram, with the highest value for the solid portion of the digestate from thermophilic anaerobic digestion	

Table 14 (continued)

Digestate supply	Thermal operation and temperature	Heating rate and residence time	Outcome (%)	Char properties	References
Solid food waste	Pyrolysis at 400–800 °C	10 °C/min; 2 h	36.13–44.26	Yield decreased with increased temperature Higher ash percentage of 34.27–37.92 weight% and fixed carbon (29.49–48.25%) with increased pyrolysis temperature Surface areas increased from 4.74 to 462.83 square metres/gram with increased pyrolysis temperature Nitrogen, hydrogen, and oxygen are reduced with increased pyrolysis temperature	Liu et al. (2020b)
Swine manure	Pyrolysis at 300–900 °C	10 °C/min; 0.5 h	Not mentioned	Lower heating value than raw digestate The heating value declined with rising temperature Lowering surface area up to 700 °C, with the highest of 102 square metres/gram at 800 °C Alkali/alkaline earth element enhanced	Hung et al. (2017)

Table 14 (continued)

Digestate supply	Thermal operation and temperature	Heating rate and residence time	Outcome (%)	Char properties	References
Agricultural residues and herbaceous biomass	Pyrolysis at 500 °C;1 h	5 °C/min; 1 h	33.1	Carbon and ash contents increased to 64.3% and 24.60%, respectively, than the raw digestate and 62.85% and 12.30% for hydrochar, respectively. Low specific surface areas (23.10 square metres/gram). Lower average pore diameter (14.80 nm) than hydrochar (16.50 nm). Lower oxygen/carbon (0.07) and hydrogen/carbon (0.50) ratios than hydrochar (0.2 and 1.06, respectively)	Miliotti et al. (2020)
	Hydrothermal carbonisation at 200 and 250 °C	0.5–3 h	51.0–72.6	Yield reduced with increased temperature. Higher ash and carbon (maximum 62.85) contents than the raw digestate. Nitrogen content is positively proportional to the temperature and negatively with the time (maximum 1.69%). Low surface areas (4.9 square metres/gram)	

Table 14 (continued)

Digestate supply	Thermal operation and temperature	Heating rate and residence time	Outcome (%)	Char properties	References
Cow manure	Hydrothermal carbonisation at 180–240 °C	1 h	58–75	Yield is negatively proportional to pyrolysis temperature rise	Belete et al. (2021)
	Hydrothermal carbonisation at 180–240 °C	1 h; dried digestate to whey aspects of 1/3	69–87	Carbon content increased with increasing temperature (maximum is 37.7 at 240 °C, digestate + whey) Higher carbon, phosphorus, and ash contents Lower oxygen content than the digestate Nitrogen content was steady with temperature increased Higher heating value than raw digestate Net energy gain of 4.4–5.3 megajoules/kilogramme for combined hydrothermal carbonisation of digestate + whey, while digestate- hydrothermal carbonisation + water produced 2.3–2.9 megajoules/kilogramme	

Table 14 (continued)

Digestate supply	Thermal operation and temperature	Heating rate and residence time	Outcome (%)	Char properties	References
Organic household waste Cow manure Energy crops	Hydrothermal carbonisation at 170–250 °C	2 and 5 h	55.0–85.9 (dry ash-free basis) 47.3–90.6 (dry ash-free basis) 51.1–91.0 (dry ash-free basis)	Yield reduced with increased temperature Higher fixed carbon with increased temperature (the maximum increase was 34.1, 39.0, and 41.4% dry matter basis for household waste, energy crop, and cattle manure, respectively) Oxygen/carbon and hydrogen/carbon ratios are reduced as temperatures increase Higher ash content (46.8, 23.9, and 37.8%, respectively) Higher heating calorific value with time and temperature increase (maximum 20.7 megajoules/kilogramme for dry manure base)	Cao et al. (2019)
Sewage	Hydrothermal carbonisation at 250 °C	0.5 h	68–76	Yields were proportional to the solid loading rate Ash reduced with increased temperature Fixed carbon (maximum 8.4%), carbon (maximum 34.4%), and nitrogen (maximum 2.8%) content improved, while oxygen and hydrogen contents decreased with increased solid loading The heating value (maximum 16.5 megajoules/kilogramme) is higher than raw digestate (14.54 megajoules/kilogramme)	Aragón-Briceño et al. (2020)
Food waste	Gasification at 850 °C	15 min	16.78–29.74	High specific surface areas (160.6–365.26 square metres/gram) The average pore size ranged between 5.72 and 4.04 nm) Nutrient enriched (nitrogen, phosphorus, and potassium)	Zhang et al. (2021)

Table 14 (continued)

Digestate supply	Thermal operation and temperature	Heating rate and residence time	Outcome (%)	Char properties	References
Sewage sludge and food waste	Pyrolysis 550 °C at different mixing ratios with sewage sludge (3:1, 2:2, 1:3)	10 °C/min	62.71–74.86	Yields are proportional to increased food waste portion; Ash is reduced with the increase in food waste; Average pore sizes ranged from 6.5–9.7 nm; The surface area ranged from 18.74–31.3 square metres/gram	Wang et al. (2022b)
Agricultural waste	Hydrothermal carbonisation at 190 °C	30 min	Not mentioned	Fixed carbon, carbon, and oxygen increased with hydrochar than the raw digestate; The calorific value increased from 20.89 to 22.49 megajoules/kilogramme; Ash content, hydrogen, and nitrogen were slightly reduced; Oxygen/carbon and hydrogen/carbon ratios were reduced; Better fuels potentials than raw digestate	Wang et al. (2022c)
Cow manure digestate				Ash increased; Fixed carbon and carbon increased; The calorific value increased from 17.0 to 17.7 megajoule/kilogramme; Oxygen/carbon and hydrogen/carbon ratios were reduced; Better fuels potentials than raw digestate	
Municipal sewage sludge				Ash largely increased; Carbon reduced; Calorific value reduced from 17.55 to 16.74 megajoules/kilogramme); Oxygen/carbon and hydrogen/carbon ratios were reduced; Better fuels potentials than raw digestate	

Table 14 (continued)

Digestate supply	Thermal operation and temperature	Heating rate and residence time	Outcome (%)	Char properties	References
Food waste of high and low solid anaerobic digestates	Autogenic pressure carbonisation at 300 to 700 °C	5 °C/min for 30 min	49.4– 78.7	Yields reduced with the increased temperature Syngas yield (maximum 39.8%) was obtained at 700 °C Syngas heating values of 22.4–24.6 megajoules/cubic metre Char at 700 °C has a heating value of 15.07 megajoules/kilogramme at high solid loads compared to 3.76 at a lower solid rate The energy at a high solid rate was higher than the raw food digestate	Peng et al. (2022)
Food waste with different moisture content	Pyrolysis at 600 °C	20 min	33.57 to 30.19	Reduced carbon and nitrogen contents Carbon/hydrogen and carbon/oxygen ratios increased with increased moisture from 5 to 60%, whereas moisture increased reduced the biochar yields from 33.57 to 30.19% and the carbon content to 20.28–21.43%	Wang et al. (2022d)

Table 14 (continued)

Digestate supply	Thermal operation and temperature	Heating rate and residence time	Outcome (%)	Char properties	References
Food waste	Pyrolysis 160–260 °C At 2 to 25% loading rates	20–120 min at different solid loading rates (49.8–68.3%, 53.6–62.5%, and 58.1–64.6%)	68.9 to 58.6	Yields slightly reduced from 68.9% to 67.5% to 58.6%, with the temperature increase to 160, 200, and 260, respectively Yields gradually decreased when increasing the reaction time Yields increased when increasing the solid loading rate The average heating rates increased up to 94.912–106.499 kilojoules/mole compared with 85.181 kilojoules/mole from the raw food wastes	Xie et al. (2022)

Numerous thermochemical methods for producing biochar are discussed, including pyrolysis, hydrothermal carbonisation, and gasification. Pyrolysis and hydrothermal carbonisation are two processes that can significantly increase the yield of biochar from anaerobic digestate. At heating rates of less than 40 °C/min and temperatures ranging from 160 to 900 °C, a typical biochar yield of about 22–78% can be obtained using the pyrolysis mode. A temperature range of 400–650 °C is optimal for producing biochar from digestate with enhanced properties, most notably specific surface area, pore-volume, and particle size

economic load (Pecchi and Baratieri 2019). The moisture content of the digestate prior to gasification has a significant effect on both the total energy efficiency and the syngas value (Yao et al. 2017). As a result, the heat fraction produced by gasification or pyrolysis should be returned to the digestate dryness process to make the operation more viable. Otherwise, as part of an integrated biogas-thermochemical approach, biogas energy could provide dryness energy.

As an alternative to digestate drying, the hydrothermal carbonisation process is not constrained by the moisture content of the biomass, making hydrothermal carbonisation most suitable for biogas digestate with a water content greater than 80% (Song et al. 2020). This may result in a reduction in the energy required for dryness (González et al. 2021). The hydrothermal carbonisation process is carried out at temperatures ranging from 150 to 250 °C and takes between 0.5 and 5 h to complete (Miliotti et al. 2020; Belete et al. 2021; Cao et al. 2019; Wang et al. 2022c) produced 47–91% hydrochar. The calorific value of hydrochar increased from 20.89 to 22.49 MJ/kg, which ensured better potential to be used as fuels than raw digestate (Wang et al. 2022c). Hydrothermal carbonisation operated at mild temperatures can convert wet biomass, including digestate, into a non-infectious carbonaceous hydrochar (Belete et al. 2021). The combination of compressed hot water and feedstocks improves mass and heat transfer at lower temperatures than pyrolysis and gasification (Miliotti et al. 2020; Chen et al. 2017). Additionally, the solid fraction produced (hydrochars) is significantly greater than that produced by pyrolysis and gasification, as shown in Tables 13 and 14 (Miliotti et al. 2020; Chen et al. 2017). Belete et al. (2021) found that hydrochar produced from cow manure digestate contained increased phosphorus, implying that hydrochar could be used to fertilise soils deficient in phosphorus.

The hydrothermal reaction produces a minor gaseous fraction (2–5%) dominated by carbon dioxide and a moderate liquid fraction of up to 20%; thus, the liquid must be processed appropriately (Zhang et al. 2019). Additionally, industrial-scale processes require large amounts of water; thus, recirculating water has been proposed as a feedstock for anaerobic digestion in order to alleviate the process's high-water requirement (Kambo and Dutta 2015; Aragón-Briceño et al. 2020; Wang et al. 2022e). Utilising the liquid fraction as a return feedstock for the anaerobic digestion is a promising application that could yield 0.304-L methane/gram chemical oxygen demand from pine sawdust and sewage sludge (Wang et al. 2022e) and 0.146-L methane/gram chemical oxygen demand for the food waste feedstock compared to 0.06-L methane/gram chemical oxygen demand for the non-hydrochar supplemented reactors (Zhao et al. 2018). The increased methane yield is attributed to the hydrothermal reaction solubilising more organic compounds (Wirth et al. 2015).

Although hydrothermal carbonisation has the potential for valorising digestate/feedstocks, increasing the energy produced by returning the liquid fraction in anaerobic digestion and treating all feedstocks, including digestate, with high moisture contents without the need for a pre-drying phase, hydrothermal carbonisation has several disadvantages. From an energetic standpoint, hydrochar may achieve a slightly more calorific value than raw digestate due to the deoxygenation reaction, and the increased ash content of hydrochar may prevent hydrochar use as a solidified fuel (Belete et al. 2021; Aragón-Briceño et al. 2020). Given the hydrothermal carbonisation of the digestate, obtaining a liquid fraction is challenging. Additionally, the hydrochar fraction is less stable (high oxygen/carbon ratio) than biochar formed via slow pyrolysis. Thus, from the perspective of biochar carbon sequestration, biochar produced via the pyrolysis process is the optimal choice, as discussed in the following section.

Role of biochar from digestate in climate change mitigation

Carbon storage and stability Biochar is being considered a possible candidate for climate change crisis mitigation. The decrease in greenhouse gas emissions facilitates biochar carbon sequestration in the soil. Thus, biochar stability is the most critical factor determining biochar's carbon sequestration capacity.

Thermal treatment modifies the chemical properties of the carbon in biochar, resulting in aromatic assets that are highly resistant to microbial biodegradation and have a long period of stability, which could be hundreds or thousands of years. Biochar is typically composed of fixed or stable carbon, labile carbon, moisture, volatile organic compounds, and ash.

Biochar carbon stability can be detected from: (1) biochar carbon structure that reflects the degree of aromatic condensation "aromaticity ", (2) biochar oxidation resistance obtained from thermal recalcitrance index, and (3) biochar persistence from hydrogen/carbon$_{org}$ and oxygen/carbon$_{org}$ elemental ratios (Leng et al. 2019). In general, hydrogen/carbon$_{org}$ is defined as the most appropriate surrogate for biochar stability. The upper thresholds of 0.4 for oxygen/carbon and < 0.7 for hydrogen/carbon$_{org}$ are required for standard-compatible biochar. At hydrogen/carbon of maximum 0.7, ample fused aromatic ring structures are ensured, which are used to differentiate biochar from the raw biomass or other substrates that are only deficiently or partially carbonised (Leng et al. 2019). Moreover, different oxygen/carbon ratios of biochar can lead to different stability, where biochar has an oxygen/carbon molar ratio lower than 0.2 is the most stable, holding an assessed half-life of more than 1000 years; biochars with an oxygen/carbon ratio of 0.2–0.6 poses intermediate half-life of 100 to 1000 years; and, that with an oxygen/carbon ratio of more than 0.6 has

a half-life of less than 100 years (Leng et al. 2019; Spokas 2010).

On the other hand, the biochar stability is negatively related to oxygen/carbon ratios (Leng et al. 2019; Spokas 2010). Leng et al. (2019) showed that biochar with oxygen/carbon of more than 0.2 or hydrogen/carbon of lower than 0.4 has mild sequestration potential, while those of oxygen/carbon less than 0.2 or hydrogen/carbon$_{org}$ less than 0.4 present high carbon sequestration ability. Shen et al. (2020) reported that the hydrogen/carbon ratio signifies the degree of carbonisation for the biochar organic aromaticity, where a hydrogen/carbon molar ratio lower than 0.3 poses extremely condensed aromatic ring structures, while a hydrogen/carbon molar ratio more than 0.7 indicates a non-condensed aromatic structure. For example, Lorenz and Lal (2018) stated that biochar could remain stable in soil for 1000 to 10,000 years, with an average of 5000 years, without any biodegradation. Fixed carbon is strongly correlated with stable carbon content, which increases the resistance of biochar to biological decomposition (Qambrani et al. 2017).

The fixed carbon value of biochar demonstrates biochar's environmental potential as an effective negative emissions technology tool, with higher fixed carbon yields implying greater biochar potential to mitigate climate change (Brassard et al. 2016). Biochar's fixed carbon and aromatic carbon contents are generally proportional to the increase in pyrolysis temperature, implying the removal of volatile materials, and that properties can be used as a proxy for biochar stability in thermochemical mode (Kannan et al. 2017; Manyà et al. 2014). As a result, biochar is considered an effective tool for long-term carbon sequestration (Qambrani et al. 2017; Ghani et al. 2013; Qiao and Wu 2022).

A summary of the different biochar features originating from various biomass, including anaerobic digestates, compared to their original raw mass is described in Table 15.

Liu et al. (2020b) observed that increasing pyrolysis temperature decreased the pyrolytic biochar yields while increasing the ash and fixed carbon contents, thereby increasing the stability of carbon in the biochar. Miliotti et al. (2020) investigated the carbonisation capacity of slow pyrolysis (500 °C for 1 h) and hydrothermal processes (200–250 °C for 0.5–3 h) on the anaerobic digestates from agro-industrial residues and herbaceous biomass. The authors found that biochar contained slightly more carbon (64.3% weight/weight) than hydrochar (62.9% weight/weight at 250 °C for 3 h). Although the specific areas of all chars were reduced, the biochar area was significantly greater than the hydrochar area of 4.92 m^2/g at 23.10 m^2/g. They found that the biochar had lower hydrogen/carbon and oxygen/carbon molar ratios, indicating greater carbon stability and resulting in a dark grey discolouration of the biochar, as opposed to brownish hydrochar discolouration (mild hydrochar carbonisation). Besides, the authors stated that

hydrochar is comparable to sub-bituminous coal in terms of discolouration, whereas biochar is comparable to anthracite.

Additionally, biochar contained a low concentration of polycyclic aromatic hydrocarbons and a greater surface specific area. They concluded that while the oxygen/carbon$_{org}$ and hydrogen/carbon$_{org}$ molar ratios of biochar produced met the requirements of the Italian fertilisation decree and the International Biochar Initiative (hydrogen/carbon molar ratio 0.7), hydrochar exceeded the upper limit values due to hydrochar high hydrogen content. Likewise, Wang et al. (2022c) studied the carbon stability of four digestates and their hydrochars and found higher carbon stability for all hydrochars than the raw digestate. The hydrothermal carbonisation of digestate reduced the oxygen/carbon and hydrogen/carbon ratios of agriculture wastes from 0.36–1.31 to 0.35–1.22, respectively. However, the hydrogen/carbon values are more than 0.7.

Wang et al. (2022d) investigated the effect of pyrolysis of digestate obtained from food waste with varying moisture contents (5, 20, 40, and 60%). They found more mesopore and micropore structures in the biochar as the moisture content increased from 5 to 60%, causing an increase in the biochar's Brunauer–Emmett–Teller surface area from 89.23 to 117.75 m^2/g . In addition, moisture promoted the formation of oxygen-containing functional groups and amorphous carbon structures, which are advantageous for the biochar's sorption abilities. Fixed carbon, carbon/hydrogen, and oxygen/carbon have increased from 1.06%, 0.55%, and 5.08% to 10.18–13.09%, 2.17–34.30%, and 11.68–51.75%, respectively, indicating that carbon stability, improved aromaticity and decreased polarity improved the carbon sequestration potential of pyrolysed digestate. Increased moisture percentages resulted in increased carbon/hydrogen and carbon–oxygen/atomic ratios, increasing carbon stability. The increased aromaticity indicated that the biochar was more stable and resistant to microorganism decomposition (Leng et al. 2019).

Nair et al. (2020) examined biochar's carbon sequestration potential and biochar's ability to act as a long-term carbon sink in soils. Biochar was produced from three organic wastes: banana fibrous waste, sewage sludge from wastewater treatment, and anaerobic food digestate. The authors noted that biochar derived from sewage sludge and anaerobic food digestate contained higher concentrations of alkali/alkaline earth metals, ash, and chloride than biochar made from banana fibrous waste. Additionally, they found that food digestate had the highest thermal-oxidative recalcitrance index of 0.294 at 650 °C slow pyrolysis, compared to 0.278 for sewage sludge, indicating that biochars in soil have a longer carbon sink potential. During pyrolysis, anaerobic food digestates contain the highest concentrations of stable phosphorus and chloride, which may increase the digestate's oxidative stability. However, a mixture of the three substrates had the highest carbon sink potential than their

Table 15 Biochar properties from several digestates

Digestate type	Thermochemical treatment/temperature (°C)/heating rate/reaction time (h)	Biochar/hydrochar characteristics										Raw digestate characteristics									References
		pH	Fixed carbon (%)	C (%)	H (%)	O (%)	N (%)	H/C	O/C	Ash content (weight%)	Char Yield (%)	pH	Fixed carbon (%)	C (%)	H (%)	O (%)	N (%)	H/C	O/C	Ash (%)	
Food waste with moisture contents of 5–60%	Pyrolysis 600 °C/20 min	Not mentioned	10.18–13.09	20.28–21.43	0.05–0.78	0.53–2.31	2.34–3.12	2.17–34.30*	11.68–51.75*	72.27–74.09	33.57 to 30.19	Not mentioned	1.06	50.24	7.60	13.19	3.21	0.55*	5.08*	24.30	Wang et al. (2022d)
Agricultural waste	Hydrothermal carbonisation /190 °C/30 min	Not mentioned	22.50	52.8	5.40	24.66	4.31	1.22	0.35	12.20	Not mentioned	Not mentioned	19.2	50.0	5.50	23.88	4.99	1.31	0.36	14.2	Wang et al. (2022c)
Cow manure digestate		Not mentioned	25.40	45.20	4.50	30.20	2.50	1.19	0.50	17.20	Not mentioned	Not mentioned	20.20	42.60	5.0	34.30	2.0	1.40	0.60	15.70	
Municipal sewage sludge		Not mentioned	Not mentioned	35.92	5.12	7.96	1.86	1.70	0.17	46.65	Not mentioned	Not mentioned	Not mentioned	36.33	5.90	12.81	4.23	1.93	0.26	33.14	
A 42% cow manure, 28% grass silage, 20% maize silage, 9% whole plant silage, and 1% cereals (% in weight)		Not mentioned	27.10	40.50	4.0	24.10	2.10	1.18	0.45	28.90	Not mentioned	Not mentioned	21.60	40.30	4.60	24.0	2.10	1.36	0.45	28.7	
Solid food waste	Pyrolysis/400–800 °C/10 °C/min/2 h	Not mentioned	29.49–48.25	46.8–50.41	1.09–2.12	8.92–14.66	1.79–1.90	Not mentioned	Not mentioned	34.27–37.92	36.13–44.26	Not mentioned	23.69	43.52	4.84	39.4	1.92	Not mentioned	Not mentioned	10.21	Liu et al. (2020b)
Agro-industrial residues and herbaceous biomass and	Pyrolysis/500 °C/1 h/5 °C/min	9.40	58.60	64.34	2.68	6.38	1.78	0.50	0.07	24.60	Not mentioned	7.70	21.80	46.70	5.50	36.9	1.20	Not mentioned	Not mentioned	9.30	Miliotti et al. (2020)
	Hydrothermal carbonisation/200–250 °C/0.5–3 h	Not mentioned	Not mentioned	49.69–62.85	5.55–5.90	17.17	0.99–1.69	1.06	0.20	12.30											Miliotti et al. (2020)
Digestate feedstock	Hydrothermal liquefaction/350–550 °C/5 °C/min	Not mentioned	Not mentioned	13.57	1.89	7.85	0.89	0.14	0.02	75.47	0.89	Not mentioned	Not mentioned	28.64	4.45	24.6	2.8	~0.16	~0.9	39.53	Okoro and Sun (2021)
Food waste digestate	Microwave-assisted hydrothermal carbonisation/160–260 °C/20–120 min	Not mentioned	Not mentioned	~47–68	~8	~19–39	~4	1.84–1.29	0.57–0.24	Not mentioned	Not mentioned	Not mentioned	26.9	47.34	8.23	39.57	4.86	2.1	~6.4	~4.7	Xie et al. (2022)

Biochar/hydrochar production increased the carbon contents, while the hydrogen/carbon (H/C) and the oxygen/carbon (O/C) molar ratios were reduced, verifying better stability than the raw digestates. The hydrogen/carbon ratio reduced from 0.16–2.1 to 1.84–0.13. Overall, hydrogen/carbon$_{org}$ and oxygen/carbon$_{org}$ are the most appropriate proxy for biochar stability. The upper thresholds of 0.4 for oxygen/carbon and <0.7 for hydrogen/carbon$_{org}$ are required for standard-compatible biochar. *The ratios are estimated as carbon/oxygen and carbon/hydrogen. C: Carbon. H: Hydrogen. N: Nitrogen. O: Oxygen

individual potential (Nair et al. 2020). As a result, biochar-derived digestate has the potential to be an efficient carbon sink during carbon dioxide removal.

Carbon sequestration and greenhouse gases mitigation The carbon sequestration process captures and stores carbon to avoid atmospheric emissions (Osman et al. 2021a). The transfer of carbon into an inert or stable passive pool to decrease carbon emission is essential. Biochar can provide a simple way to shift the carbon from an active to a passive pool (Fawzy et al. 2021). As previously discussed, biochars are more chemically and biologically stable than the initial carbon forms; thus, their release as carbon dioxide is difficult, making biochar a promising carbon sequestration tool (Fawzy et al. 2021).

Transferring small amounts of carbon, such as 1% of the net annual carbon cycled between the atmosphere and plants, to biochar would significantly reduce approximately 10% of current carbon emissions from anthropogenic sources, as annual atmospheric carbon dioxide uptake by plants via photosynthesis is nearly eight times greater than anthropogenic greenhouse gas emissions. Thus, some authors estimated that annually producing 3 gigatonnes of biochar (from 60.6 gigatonnes of biomass pyrolysis) would prevent approximately 3 gigatonnes of carbon from being emitted into the atmosphere (Qambrani et al. 2017; Sri Shalini et al. 2021). By 2030, approximately 1 gigatonne of carbon will be sequestered annually, which meets the biochar potential. Additionally, biochar production, storage, and use as a carbon-negative source would sequester between 0.3 and 2 gigatonnes of carbon dioxide per year by 2050 (Fawzy et al. 2021).

Alhashimi and Aktas (2017) compared the global warming potential of numerous biochars derived from poultry litter, cattle manure, agriculture, sewage sludge, and food waste. They reported that biochar has a carbon negative emission of -0.9 kg carbon dioxide equivalent per kilogramme on an average basis, owing to biochar carbon sequestration capacity, compared to 6.6 kg carbon dioxide equivalent per kilogramme of activated carbon. Additionally, the energy required to form 1 kg of biochar and activated carbon was 6.1 MJ/k and 97 MJ/k, respectively. However, more calculations are still required based on transportation cost, economic performance, adsorption cost, and environmental impact.

Song et al. (2020) evaluated the sewage sludge lifecycle by incorporating anaerobic digestion and fast pyrolysis compared to fast pyrolysis only. The net greenhouse gas emissions for the integrated process were -15.8 tonnes of carbon dioxide equivalents compared to -11.8 tonnes of carbon dioxide equivalent for fast pyrolysis alone. This indicated that less greenhouse gas should be emitted than consumed in order to achieve a beneficial climate change effect.

Likewise, Zhao et al. (2018) stated that biomass pyrolysis could be a viable alternative to industrial coal-fired boilers because biomass pyrolysis produces biochar, bio-oil, and syngas at a lower temperature. According to the authors, the biomass-based pyrolysis Hubei Pyrolysis plant in China has the potential to emit zero net greenhouse gases if 41.02% of the biochar is applied to the soil while utilising all of the biochar in the field could save -32 million tonnes of carbon dioxide equivalent.

Lin et al. (2021) simulated two systems for converting carbon dioxide to biomethane and digestate to biochar for carbon dioxide sequestration via pyrolysis in conjunction with a conventionally operating biogas plant. The authors proposed carbon capture and utilisation using a cascading circular approach involving microbial electrolysis cells, power to gas conversion, and digestate valorisation for biochar formation. The efficacy of the cascading circular bioenergy system was demonstrated using cattle slurry as a feedstock in three study areas, including the European Union, China, and the United States of America. Annual greenhouse gas emission savings were calculated using 1.09, 0.78, and 1.16 billion tonnes of cattle slurries produced in the European Union, China, and the United States, respectively. The authors calculated that using renewable electricity to power microbial electrolysis cells would result in annual greenhouse gas emissions savings of 397.4 million tonnes of carbon dioxide equivalent, or 150.1, 94.2, and 153.1 million tonnes of carbon dioxide equivalent for the three regions, respectively. However, suppose grid electricity is used to power microbial electrolysis cells. In that case, annual greenhouse gas emissions savings total 159.2 million tonnes of carbon dioxide equivalent, with the European Union contributing 102.2 million tonnes of carbon dioxide equivalent, China contributing -10.1 million tonnes of carbon dioxide equivalent, and the United States contributing 67.0 million tonnes carbon dioxide equivalent. The system combining microbial electrolysis cell and anaerobic digestion has the potential to generate 2.29 exajoules of total energy or about 2% of global natural gas consumption in 2018. Meanwhile, the systems demonstrated a capacity for negative carbon emissions via biochar generation, with biochar application capable of reducing annual greenhouse gas emissions by 2% (totalling 20.1 gigatonnes carbon dioxide equivalent) in the three regions, as shown in Table 16.

Utilisation of biochar for biogas upgradation

The raw biogas produced by anaerobic digestion is primarily composed of methane (50–70%) and carbon dioxide (30–50%), with trace amounts of ammonia, water vapour, hydrogen sulphide, and oxygen and nitrogen (Angelidaki et al. 2018). Cleaning and upgrading biogas are necessary to meet grid and engine requirements; however, biogas

Table 16 Carbon capture and utilisation by integrating microbial electrolysis cell, power to gas, and digestate valorisation for biochar production (Lin et al. [2021])

Parameters	Conventional biogas plant	Biogas plant with digestate pyrolysis	Biogas plant with microbial electrolysis and digestate pyrolysis
Feedstock rate	100 tonnes of wet cattle slurry/day with 8.75 tonnes and 6.69 tonnes of total solids and volatile solids, respectively		
Slurry production/year	36,500 tonnes/year from 3000 cattle (12.3 tonnes slurry/head/year)		
System	Biogas production and upgrading		
Greenhouse gases emission from digestate management	Not included		
Pyrolysis products	Not included	Biochar, syngas, and bio-oil	Biochar, syngas, and bio-oil
Biogas upgrading	Amine scrubbing	Biological through biochar addition	Biological through biochar addition
Biomethane production	1.14 tonnes of biomethane (15.99 cubic metres methane/tonne wet weight), 60% methane content	2.03 tonnes (equal to 28.46 cubic metres methane/tonne wet weight, a 78.1% improvement over the traditional scenario) and 0.01-tonne hydrogen	2.28 tonnes (comparable to 31.87 cubic metres methane/tonne wet weight, a 100% improvement over the traditional scenario)
Other products	0.13 tonnes of carbon dioxide and 15.56 tonnes of solid digestate	0.32-tonne syngas, 0.96-tonne bio-oil, and 1.20 tonnes biochar	0.32-tonnes syngas, 0.96-tonne bio-oil, and 1.20 tonnes biochar
Hydrogen production	Not included	Electricity	Microbial electrolysis
Greenhouse gases saving	7.28 and 8.23 tonnes carbon dioxide equivalent/100 tonnes of cattle slurry/day	9.68–12.38 tonnes carbon dioxide equivalent/100 tonnes of feedstock/day	12.07–13.20 tonnes carbon dioxide equivalent/100 tonnes of feedstock/day

Two systems are modelled in conjunction with a conventional running biogas plant to convert carbon dioxide to biomethane and digestate to biochar via pyrolysis. One system incorporates a biogas plant and digestate pyrolysis, while another incorporates a biogas plant and microbial electrolysis. The latter model demonstrated the greatest potential for carbon dioxide sequestration, at 12.07–13.20 tonnes of carbon dioxide equivalent/100 tonnes of feedstock per day. Both systems saved more carbon than a conventional biogas system without digestate pyrolysis. The overall reduction in greenhouse gas emissions was attributed to (1) carbon dioxide saved by substituting biomethane and biochar by-products such as syngas and bio-oil for fossil diesel fuels, (2) avoided methane emissions from traditional manure storage, and (3) carbon sequestration potential through the use of solid digestate-driven biochars as a long-term soil amendment agent

purification steps can account for up to 55% of the total cost of biomethane generation (Chiappero et al. 2020; Shen et al. 2016). Until now, conventional technologies such as scrubbings, chemical absorption, cryogenic separation, and membranes have been used extensively. Recently, biochar has been investigated as an in situ and ex-situ adsorbent for hydrogen sulphide and carbon dioxide. The gaseous adsorption capacity of biochars is determined by inherited physicochemical properties such as porosity, alkalinity, hydrophobicity, the presence of surface functional groups, and aromaticity (Dissanayake et al. 2020).

Acid-basic reactions between acidic carbon dioxide and the alkaline biochar surface promote carbon dioxide adsorption (Lahijani et al. 2018; Saha and Kienbaum 2019). Chemical modifications to increase the alkalinity of biochar would be more beneficial for increasing the selectivity and adsorption of carbon dioxide (Lahijani et al. 2018; Zhou et al. 2017). The sorption of hydrogen sulphide from biogas to the alkaline surface of biochar followed a similar pattern (Sahota et al. 2018b).

Role of biochar in "in situ" biogas upgrading Several studies (Shen et al. 2016; Shen et al. 2017; Linville et al. 2017) explored the viability of in situ biogas upgradation by biochar addition to ensure that the obtained biomethane contents matched the quality of pipelines.

Shen et al. (2015) investigated the possibility of carbon dioxide sequestration via biochar addition to waste activated sludge thermophilic anaerobic digestion. The in situ biogas upgrading process resulted in an average methane content of 88.5–96.7% in biochar-supplemented bioreactors, compared to 67.9% in a control bioreactor. Additionally, carbon dioxide was removed at a rate of 54.9–86.3%, with a residual hydrogen sulphide concentration of less than 5 parts per billion obtained. The authors suggested that the large surface area, high porosity, the abundance of basic structures, and more hydrophobic sites of biochars could support carbon dioxide mitigation. Likewise, Shen et al. (2016) investigated the effect of two woody biochars on the biogas upgrading process in anaerobic digestion. They observed increased methane contents of up to 92.3% and 79.0% in reactors amended with mesophilic and thermophilic biochar, respectively, corresponding to 66.2% and 32.4% carbon dioxide removal, respectively. The carbon dioxide sequestration capacity of the biochars used was attributed to inherited biochar characteristics such as high specific surface area, chemical stability, porosity, alkaline nature, and degree of carbonisation. Nevertheless, Shen et al. (2016); Shen et al. (2015) observed a decrease in methane productivity with increasing biochar dosages, concluding that biomethane inhibition may be caused by the increased cations released by the increased biochar dosages.

In two-stage reactors, Shen et al. (2017) examined the effect of pine wood and corn stover biochars on the anaerobic digestion of waste activated sludge. They recorded an average methane content of 81.0–88.6% in the digester supplied with corn stover biochar and 72.1–76.6% in the digester supplied with pine wood biochar, compared to approximately 70.0% for the non-amended control digester. They confirmed that biochars could sequester carbon dioxide through chemical adsorption and the formation of carbonate/bicarbonate salts facilitated by the biochar surface structure. Linville et al. (2017) studied the effect of walnut shell-originated biochar's particle size and dosage on food waste anaerobic digestion under mesophilic and thermophilic conditions. The authors observed greater carbon dioxide removal in the bioreactors amended with smaller particle-sized biochar (61%) than in those amended with larger particle-sizes biochar (51%), owing to the larger surface areas and ash contents. Few authors have confirmed the beneficial role of in situ desulphurisation of biochar; for example, Choudhury and Lansing (2020) reported that the hydrogen sulphide content in biogas was decreased from above 1500 parts per million to less than 160 parts per million after amending maple wood or corn stover biochars at a rate of 1.82 g/g total solids of dairy manure under mesophilic batch digesters.

Apart from the adsorption pathway for carbon dioxide on biochar, almost methane formation relies on the syntrophic interspecies electron transfers between organic acid-oxidising bacteria and carbon dioxide-reducing methanogens (Masebinu et al. 2019; Pan et al. 2019). Yang et al. (2020) investigated the biomethanation of carbon dioxide through three carrier materials for hydrogenotrophic methanogens, which involved commercial black ceramsite, corn straw biochar, and digestate biochar. They demonstrated that the digestate biochar option had the highest methane rate of 0.345 L/l$_{reactor}$/day, which was 10.7 folds higher than that of the black ceramsite group. In addition, the hydrogen utilisation rate of the digestate biochar system was also the highest (1.18 L/l$_{reactor}$/day). The higher biomethane generation was attributed to the role of biochar surface as a biofilm carrier material for immobilisation of hydrogenotrophic methanogens, which led to higher microbial densities, faster metabolism, and more microbial stability, ensuring better conversion of carbon dioxide into methane. Further approval by other studies about in situ biogas upgradation by biochar would be valuable.

The in situ supplementation of biochar in the anaerobic digestion bioreactors can protect the microbial communities from excessive hydrogen partial pressure, a most commonly technical limitation observed during in situ biogas upgradation via hydrogenotrophic methanogenesis way (Fu et al. 2021; Agneessens et al. 2017). In addition, decreased bulk density of biochar could improve the hydrogen mass transfer between gas and liquid phase (D'Silva et al. 2021),

Table 17 Biochar sorption ability of hydrogen sulphide and carbon dioxide during ex-situ functions

Feed materials	Thermal operation	Pore volume (cubic centimetre/gram)	Surface area (square metre/gram)	Gas	Inlet gas flow (part per million)	Hydrogen sulphide adsorption	Carbon dioxide adsorption	References
Anaerobic digestate fibres	Pyrolysis: 500 °C for 60 min	0.037	134	Synthetic biogas	2000	135 mg/g	Not mentioned	Pelaez-Samaniego et al. (2018)
	Pyrolysis: 600 °C for 60 min	0.035	142	Synthetic biogas	2000	542 mg/g	Not mentioned	
	Pyrolysis: 600 °C for 60 min + sodium carbonate	Not mentioned	Not mentioned	Synthetic biogas	2000	652 mg/g	Not mentioned	
Black liquor biomass	Pyrolysis: 450 °C for 6 min	Not mentioned	60	Nitrogen + hydrogen sulphide	1000	70 mg/g	Not mentioned	Sun et al. (2016)
Potato peel waste	Pyrolysis: 500 °C for 5 min		63	Nitrogen + hydrogen sulphide	1000	53 mg/g	Not mentioned	Sun et al. (2017)
Wood chips (80%) & anaerobic digestion digestate (20%)	Pyrolysis: 600 °C	Not mentioned	Not mentioned	Biogas	1020	273.2 mg/g	Not mentioned	Kanjanarong et al. (2017)
Perilla leaf	Pyrolysis: 700 °C	0.1	473.4	Synthetic biogas	Not mentioned	0.537 mmol/g	2.312 mmol/g	Sethupathi et al. (2017)
Korean oak	Pyrolysis: 400 °C	0.1	270.8	Synthetic biogas	Not mentioned	0.178 mmol/g	0.597 mmol/g	
Japanese oak	Pyrolysis: 500 °C	0.2	475.6	Synthetic biogas	Not mentioned	0.167 mmol/g	0.379 mmol/g	
Soybean stover	Pyrolysis: 700 °C	0.2	420.3	Synthetic biogas	Not mentioned	0.308 mmol/g	0.707 mmol/g	
Sawdust	Gasification: 850 °C	0.0036	182.04	Nitrogen/carbon dioxide	Not mentioned	Not mentioned	470 mg/g	Madzaki and KarimGhani (2016)
Sawdust	Gasification: 850 °C + monoethanolamine	0.0070	3.17	Nitrogen/carbon dioxide	Not mentioned	Not mentioned	430 mg/g	
Whitewood	Pyrolysis: 500 °C + steam activation	0.55	840	Helium/carbon dioxide	30% mole	Not mentioned	59 mg/g	Shahkarami et al. (2015)
Whitewood	Pyrolysis: 500 °C + carbon dioxide activation	0.45	820	Helium/carbon dioxide	30% mole	Not mentioned	63 mg/g	
Whitewood	Pyrolysis: 500 °C + potassium hydroxide activation	0.62	1400	Helium/carbon dioxide	30% mole	Not mentioned	78 mg/g	
Walnut shell	Pyrolysis: 900 °C for 1.5 h	0.198	397	Not mentioned	Not mentioned	Not mentioned	72.6 mg/g	Lahijani et al. (2018)
Pig manure	Pyrolysis: 500 °C for 4 h	0.044	31.57	Nitrogen/carbon dioxide	Not mentioned	Not mentioned	23.5 mg/g	Xu et al. (2016)
Wheat straw	Pyrolysis: 500 °C for 4 h	0.041	20.20	Nitrogen/carbon dioxide	Not mentioned	Not mentioned	34.4 mg/g	
Sewage sludge	Pyrolysis: 500 °C for 4 h	0.022	10.12	Nitrogen/carbon dioxide	Not mentioned	Not mentioned	18.2 mg/g	

Various biochars show good adsorption capacity towards carbon dioxide and hydrogen sulphide. Evidently, the adsorption capability for both carbon dioxide and hydrogen sulphide is found in the wide range of 18.2–470 mg/g and 53–652 mg/g, respectively. The key pathway for carbon dioxide sequestration by biochar is physical sorption, whereas the sorption of hydrogen sulphide involves several chemical mechanisms with biochar's surface. Hence, high specific surface area, sufficient pore volume, and pore size are critical for the physical sorption of carbon dioxides, and porosity, ash, or aromatics are important factors for the chemical sorption of hydrogen sulphides

which is typically restricted in the in situ hydrogenotrophic methanogenesis as discussed earlier. Therefore, incorporating biochar into the in situ approach can improve the hydrogen-assisted biogas upgradation technology, and this integration represents a novel concept for future research.

Role of biochar in "ex-situ" biogas purification The use of biochar for carbon dioxide capturing from various gaseous streams has attracted recent attention (Dissanayake et al. 2020; Singh et al. 2019; Ahmed et al. 2020), as biochar can significantly reduce the energy resource and capital cost requirements. Studying ex-situ functions of biochar for upgrading and cleaning biogas is summarised in Table 17. Sethupathi et al. (2017) evaluated the sorption of methane, carbon dioxide and hydrogen sulphide in a simulated biogas stream by four biochars in fixed-bed adsorbers' continuous experiments. The authors described that carbon dioxide and hydrogen sulphide were captured by biochars, which demonstrated sorption capacities of up to 0.537 mmol/g for hydrogen sulphide and 2.312 mmol/g for carbon dioxide. The key pathway for carbon dioxide sequestration by biochar is physical sorption; hence high specific surface area, sufficient pore volume, and pore size (0.5–0.8 nm) were extremely important (Creamer and Gao 2016).

Additionally, carbon dioxide adsorption may be influenced by the chemical characteristics of biochar, such as the presence of basic surface functional groups or alkaline and alkali earth metals, polarity, and hydrophobicity (Dissanayake et al. 2020). For instance, Xu et al. (2016) reported that the carbon dioxide adsorption by three biochars during batch tests was attributed to the occurrence of alkali and alkaline earth metals (iron, calcium, magnesium, and potassium) and carbon dioxide reactions with biochar's physical adsorption (Xu et al. 2016). Stimulation and surface modifications of biochars can support micropores and high surface area for physical adsorption and enrich surface functional groups for better chemical adsorption, causing significant carbon dioxide sorption capacities (5.0–7.42 mmol/g) (Jung et al. 2019).

Other reports used biochar to eliminate hydrogen sulphide from biogas. Sahota et al. (2018b) achieved a hydrogen sulphide removal rate of 84.2% from biogas using biochar derived from leaf waste. Kanjanarong et al. (2017) attained a 273.2 mg/g removal of hydrogen sulphide (98%) from biogas with biochar, indicating that hydroxide and carboxylic groups are accountable for hydrogen sulphide sorption. Han et al. (2020) utilised macroalgae-originated biochar (enteromorpha and sargassum) for hydrogen sulphide removal from a synthetic gas containing hydrogen sulphide (200 mg/l). The authors noticed that under 5% moist conditions, a breakthrough time was decreased from 10 to 7.4 min. However, in a dry condition, hydrogen sulphide ionisation with water produced HS⁻, which was inhibited in a moist condition due to water film occurrences in the biochar surface. The

authors also observed that at lower operating temperature (around 25 °C) and higher hydrogen sulphide flow concentration (up to 1500 ppm), biochar increased the removal of hydrogen sulphide with 0.65 mg/gram adsorption capacity (Han et al. 2020).

Sahota et al. (2018b) evaluated the viability of biochar from leaf waste to sulphurise raw biogas. Biochar produced at 400 °C demonstrated hydrogen sulphide removal of approximately 201 mg/l (84.2%) from an inlet flow of 1254 mg/l. They concluded that the biochar's increased pore size, surface area, and surface functional properties enhanced hydrogen sulphide sorption.

Das et al. (2019) removed hydrogen sulphide gas using a bio-filter filled with mature compost mixed and biochar (25% volume/volume). They found that the highest removal capacity of 33 g/cubic metre/hour was 42% greater than that of compost packing biofilter without biochar. Ultimately, Pelaez-Samaniego et al. (2018) concluded that anaerobic digestion of biochar derived from digestate could successfully eliminate hydrogen sulphide from synthetic biogas due to the existence of porosity, ash, or aromatics in biochar. Contrary to carbon dioxide, whose adsorption onto biochar appeared to be predominantly physical, the sorption of hydrogen sulphide involves several chemical mechanisms with biochar's surface (Bamdad et al. 2018).

Apart from removing hydrogen sulphide and carbon dioxide, biochar has the potential to remove other impurities from biogas, such as siloxanes. Siloxanes are compounds with "Si–O–Si" bonds that are found in anaerobic digestion feedstocks such as municipal wastes as a result of the intake of silicon-containing materials (Nyamukamba et al. 2020). Because siloxanes are insoluble in water, they are frequently affixed to sludge mass, where the larger molecules may degrade into minor particles or volatile substances that are then released into raw biogas (Cabrera-Codony et al. 2014). In raw biogas, siloxanes are difficult to remove and have the most detrimental effect on biogas utilisation (Nyamukamba et al. 2020; Piechota 2021). For instance, biogas utilisation in solid fuel cells is restricted by the production of silicon dioxides, which obstructs bioanode pores (Papurello et al. 2018). Additionally, because of biogas combustion, the silicon dioxides formed by siloxanes oxidation precipitate in engine components, causing damage to the gear and reducing equipment performance. As a result, siloxanes must be removed from biogas to improve upstream biogas applications. The adsorption concept was chosen in this instance due to adsorption's ease of operation and low cost (Nyamukamba et al. 2020; Shen et al. 2018).

The activated carbon is the frequently used siloxane sorption material from biogas for refining technology (Nguyen et al. 2021); however, biochar may be a good alternative material due to biochar enhanced physicochemical characteristics. For instance, wood waste-originated biochar

removed 3.5 to 4.4 mg/g of octamethylcyclotetrasiloxane. Nevertheless, the same authors suggested chemical or physical methods for biochar stimulation to enhance biochar sorption capacity (Papurello et al. 2019). Pristine biochar derived from coconut shells was found to be able to adsorb 223.3 mg/g of hexamethyldisiloxane, which was increased to 356.4 mg/g when biochar was loaded with 3.0% iron oxides. The adsorption capacity of biochar-loaded iron was effectively related to enhanced surface area and pore volume after metal-biochar incorporation (Meng et al. 2021). As a result, biochar can adsorb siloxanes impurity, indicating the enormous potential of engineered biochars for biogas cleaning.

In general, biochar has the potential to upgrade and clean raw biogas; however, the operating characteristics of this method require additional evaluation to ensure adequate removals. Utilising biochar in real-world, large-scale biogas systems requires consideration of the biogas composition, characteristics, operational conditions, and biochar. Thus, additional research in this area is required before scaling up a practical process.

Benefits of the circular integration of biochar with biogas system upgradation

The life cycle assessment is a technique for compiling and evaluating a material's inputs, outputs, and potential ecological effects over material useful life (Opatokun et al. 2017; Rajendran and Murthy 2019). As a result, the environmental viability of the process can be determined by integrating anaerobic digestion with the thermochemical process and subsequently utilising the biochar mass-produced from the digestate.

A life cycle assessment determined that incorporating anaerobic digestion and pyrolysis could be viable for increasing energy production and nutrient reuse by producing biogas for bioenergy and biochar for soil amendment materials. Additionally, the various aspects of biogas utilisation, such as power generation, electricity generation, household cooking, and transportation fuel, have a significant impact on the results of life cycle assessments (Mohammadi et al. 2019). The life cycle assessment of pulp and paper mill sludge was conducted under three distinct biogas end-use scenarios involving the use of biogas for vehicle fuel, electricity, heat generation, and household cooking. The life cycle assessment results indicated that integrating biogas and pyrolysis significantly reduced gas emissions under these three scenarios, owing to the dewatering and drying of the sludge and the application of biochar to the soil, which significantly reduced global warming by sequestering carbon in the field. Thus, integrating anaerobic digestion and digestate pyrolysis processes could result in a novel energy/biochar technology that maximises energy production and nutrient recovery (Mohammadi et al. 2019).

Through a life cycle analysis of the organic fraction of municipal solid waste, the effects of anaerobic digestion alone, pyrolysis alone, anaerobic digestion–pyrolysis, and pyrolysis–anaerobic digestion were evaluated on global warming, ozone depletion, eutrophication, acidification, and ecotoxicity. The integration of anaerobic digestion and pyrolysis had the lowest overall environmental impact (-11.53 total environmental effect/kilogramme organic fraction of municipal solid waste) compared to other scenarios (-8.11 for anaerobic digestion, 0.64 for pyrolysis, and 2.75 for pyrolysis-anaerobic digestion). The combination of anaerobic digestion and subsequent digestate pyrolysis resulted in the greatest environmental benefit, owing to the reduction in emissions from solid digestate landfilling and the increased production of heat and electricity for the system (Wang et al. 2021c).

Li and Feng (2018) assessed the life cycle of integrating anaerobic digestion with pyrolysis. They demonstrated better energy efficiency and environmental performance from the integration scenario than that of a single technology. A similar conclusion was obtained from anaerobic digestion-pyrolysis integration of food waste due to the utilisation of digestate-driven biochar as a fertiliser that induced the highest climate change mitigation option and better nutrients and water retentions biogas generation, and bio-oil for electricity generation (Opatokun et al. 2017).

On food wastes, life cycle assessments of integrating anaerobic digestion and gasification revealed a lower emissions approach than digestate incineration (Tong et al. 2018). Similarly, combining anaerobic digestion and hydrothermal carbonisation resulted in increased energy recovery and a 75% reduction in global warming impact, from 72 to 18 kilogrammes of carbon dioxide equivalent per tonne of sludge (Medina-Martos et al. 2020). However, challenges include lower biochar yields from gasification and decreased char stability from hydrothermal carbonisation. Thus, integration via pyrolysis would be the optimal choice.

In summary, integrating anaerobic digestion and pyrolysis to convert the digestate into value-added biochars may be a beneficial environmental strategy for reducing global warming, increasing biogas production, and curing the digestate while also producing other by-products such as bio-oil and syngas.

Biochar certification

The diversity of feedstocks, manufacturing processes, post-production processing, and the possibility of contamination all point to the importance of characterising biochar for subsequent applications. As a result, certain guidelines have been introduced to ensure that biochar possesses the characteristics necessary for various applications. The European

biochar certificate and the International Biochar Initiative are two widely recognised biochar certifications globally. The European biochar certificate established several standards for the sustainable generation of biochar in Europe, including biochar applications for European biochar certificate-Feed, European biochar certificate-Agro, European biochar certificate-Material, and European biochar certificate-AgroBio. Each category has specific requirements to ensure that biochar meets the application's requirements and that consumers receive a consistent biochar quality.

The certification process includes an assessment of the suitability of feedstock and production, sampling procedures, labelling and quality control processes, compliance with safety and health regulations, and biochar characteristics. The characteristics of biochar must adhere to the regulatory requirements for each of the specified categories. Additionally, certification requires certified analytical methods. In addition, the European biochar certificates have established standards for the biochar-based carbon sink (Fawzy et al. 2021). Table 18 summarises the regulation's requirements and limits for the specified biochar characteristics defined by the European biochar certification.

The international biochar initiative developed guidelines for characterising biochar as a soil amendment material in order to ensure consistent product quality and to inform consumers about biochar's physicochemical properties. Table 18 compares the European biochar certification to the international biochar initiative standards. Recently, anaerobic digestion digestate was added to the list of biomasses that can be used to make biochar. The legalisation specified that the biogas plant's animal feedstock must be less than 40%. Additionally, plastic contaminants must not exceed 1% in the digestate, with a 10% threshold for European biochar certificate-BasicMaterials and European biochar certificate-ConsumerMaterials. Only digestate derived from agricultural biomass is approved for the European biochar certificate-AgroOrganic (EBC 2022).

Approach and prospects

The anaerobic digestion process is based on a single conversion of feedstocks to produce biogas and digestate. Integrated biorefineries, on the other hand, can utilise additional feedstocks and may be expanded in future applications beyond anaerobic digestion to produce a variety of biofuels, power generation, and chemical materials. In this regard, biochar production can significantly alleviate the difficulties typically encountered in the biogas sector via the following trade-offs between biogas and biochar technologies:

- Solid digestate would be suitable for an effective trade-off approach between biochar and biogas technologies. The solid digestate fraction can be converted to biochar

through thermochemical methods, most notably pyrolysis. The generated biochar can be used directly as a soil amendment fertiliser or indirectly to enhance and upgrade biogas, as presented in Fig. 7.

- The direct use of digestate-derived biochar as a biofertiliser represents an interesting prospect for achieving slow-release organic fertilisers. Biochar organic biofertiliser has the potential to expand markets, overcome the huge liquid digestates produced by biogas systems, be more easily stored, less polluting, improve soil carbon stability, and help mitigate global warming. The biochar produced can be added to the open manure storage pit to reduce odorous emissions while also adsorbing nutrients from the liquid manure. On the other hand, supplementing crops silage with biochar is an intriguing strategy because that biochar has the potential to stimulate lactic acid fermentation in silage, reduce fungus formation, and reduce the risk of clostridia infections due to increased fatty acid production during silage.
- Biochar's role in anaerobic digestion's biological methanation process is extremely promising. Biochar has recently demonstrated an interest in in situ biomethane upgradation and impurity cleaning via biochar's unique properties and the recently recognised direct interspecies electron transfer between syntrophic microbes, which would benefit hydrogen-assisted pathways. Equally interesting would be biomethanation using the syngas generated during the pyrolysis process. This issue is still in its infancy, and syngas application on a larger scale is necessary. Similarly, research into bio-oil as a supplement to anaerobic digestion technology requires additional research.
- Biochar has the potential to be an efficient adsorbent for ex-situ biogas upgrading and cleaning applications. Additional research would focus on the adsorption of hydrogen sulphide, carbon dioxide, and ammonia from real biogas, taking into account their competitive sorption properties, as well as the effect of water vapour and the subsequent improvement of methane. While integrating biochar with biogas upgrading technologies, particularly membrane separation and pressure swing adsorption, which require pretreatment of hydrogen sulphide, is a promising approach, biochar may be ineffective when using moist-pressurised water scrubbing.
- The use of certain biochar-derived digestate as animal feed additives is another subsidiary option. For example, adding biochar as a feed additive can improve feed efficiency, and animal health, along with reducing greenhouse gas emissions and nutrient losses.
- Biochar-derived digestate can be combined with compost to improve the quality of the composting process in a variety of ways, including adjusting the physicochemical properties of the compost, enhancing organic matter

Table 18 Biochar properties and threshold requirements defined by the European biochar certification and the international biochar initiative (IBI 2015; IBI-EBC 2014)

| Assessment classification	Parameter/unit	International biochar initiative test procedure	International biochar initiative Standards	European biochar certification test method	European biochar certification standards
Mandatory Assessment Category A: Fundamental utility assets	Moisture content/Percentage of total mass & dry basis	American Society for Testing and Materials D1762-84: Standard method for chemical analysis of wood charcoal (require measurement date and production time). The moisture content was at 105 °C for 2 h	Declaration needed	Deutsches Institut fur Normung (DIN) 51,718 method A	Declaration needed (% of total mass, dry basis)
	Organic carbon (C_{org})/Percentage of total mass and dry basis	Total carbon and hydrogen analysis by a dry combustion-elemental analyser. Inorganic carbon measurement by detecting carbon dioxide–carbon value with 1 normal hydrochloric acid, as defined in American Society for Testing and Materials D4373 standard test procedure for rapid determination of carbonate content of soils. Organic carbon is estimated as total carbon—inorganic carbon	Minimum 10% Class 1: ≥60% Class 2: ≥30% and <60% Class 3: ≥10% and <30%	Total carbon, hydrogen, and nitrogen analysis by dry combustion-infrared detection (DIN 51,732, ISO 29541) Inorganic carbon analysis by detecting carbonate-carbon content with hydrochloric acid, as defined in DIN 51,726, ISO 925 Organic carbon is calculated as total carbon – inorganic carbon	Biochar≥ 50% Bio Carbon Minerals < 50%
	Hydrogen to organic carbon/molar ratio	Not required	The upper limit of 0.7	Same as hydrogen and organic carbon determination	0.7 maximum
	Oxygen/organic carbon/molar ratio		Not applicable	Oxygen is calculated from ash content, carbon, hydrogen, nitrogen, and sulphur (DIN 51,733, ISO 17247)	0.4 maximum
	Total nitrogen/Percentage of total mass and dry basis	Dry combustion-elemental analyser as the same technique mentioned above for the total carbon and hydrogen	Declaration needed	Dry combustion- infrared detection following the same procedure for total carbon and hydrogen (DIN 51,732)	Required (total nitrogen)

Table 18 (continued)

Assessment classification	Parameter/unit	International biochar initiative test procedure	International biochar initiative Standards	European biochar certification test method	European biochar certification standards
	Total phosphorus, potassium, magnesium, and calcium/ Percentage of total mass and dry basis		Optional (total phosphorus and potassium)	Digestion with Lithium metaborate at 550 °C according to DIN 51,729–11 and determination with ICP-MS according to DIN EN ISO 17294 or ICP-OES according to DIN EN ISO 11885	Required (Total phosphorus, potassium, magnesium, and calcium)
	Total ash/Percentage of total mass and dry basis	American Society for Testing and Materials D1762-84	Declaration needed	DIN 51,719, ISO 1171 or EN 14,775 – ashing at 550 °C, heating at 5 K/min to 106 °C under a nitrogen atmosphere, then at 5 K/min to 550 °C under oxygen, hold for 1 h	Declaration needed
	pH	pH analysis methods as defined in Sect. 04.11 of Test Methods for the Examination of Compost and Composting (2001) after Rajkovich et al. (2012) modification: Dilution of 1:20 biochar/deionised water (weight: volume) and equilibration at 90 min on the shaker	Declaration needed	DIN ISO 10390 with 1:5 biochar to 0.01-mol calcium chloride-solution, 60 min shaking, measuring directly in the suspension	Declaration (pH) If more than 10, the delivery slip must feature appropriate handling information
	Electrical conductivity/ deciSiemens per metre	Test Methods for the Examination of Compost and Composting Sect. 04.10, modified by Rajkovich et al. (2012)	Declaration needed	Method of the Bhatnagar–Gross–Krook (Federal quality community compost), volume 1, method III. C2 in analogy to DIN ISO 11265 Adding 1:10 water to the sample, shaking for 1 h, followed by filtration of the solution	Declaration needed
	Bulk density	Not required	Not applicable	Bulk density: DIN 51,705	Declaration

Table 18 (continued)

Assessment classification	Parameter/unit	International biochar initiative test procedure	International biochar initiative Standards	European biochar certification test method	European biochar certification standards
	Liming (specify for pH > 7/ Percentage of calcium carbonate	Association of Official Analytical Chemists 955.01 potentiometric titration on (wet) samples. Use dry weight to evaluate calcium carbonate percentage and report as per dry sample weight	Declaration needed	Not required	Not applicable
	Particle size distribution/Percentage fraction	Progressive dry sieving with 50 mm, 25 mm, 16 mm, 8 mm, 4 mm, 2 mm, 1 mm, and 0.5-mm sieves	Declaration needed	Not required	Not applicable
	Surface area/square metre	American Society for Testing and Materials D6556 "Standard Test Method for Carbon Black–total and external surface area by nitrogen adsorption"	Declaration needed	milled < 50 µm, 2 h outgassing at 150 °C, vacuum, nitrogen adsorption, multipoint Brunauer–Emmett–Teller method	Declaration (preferably higher than 150 square metres/g)
	Water holding capacity	Not required	Not applicable	Water holding capacity estimating by soaking and drying the sample (E DIN ISO 14238). Water holding capacity calculated as mass percentage of saturated and dry mass	Optional
	Volatile matter/Percentage of total mass and dry basis	American Society for Testing and Materials D1762-84 "Standard Test Method for Chemical Analysis of Wood Charcoal". Volatile matter content at 950 °C for 10 min	Optional	Thermal-Gravimetric-Analysis (TGA) using Leco TGA 701—total mass loss at 950 °C	Declaration needed
Mandatory Toxicant Assessment Category B: Maximum Allowed Thresholds	Germination inhibition assay	Organisation for Economic Co-operation and Development methodology (1984)	Pass/fail	Not required	Not applicable

Table 18 (continued)

Assessment classification	Parameter/unit	International biochar initiative test procedure	International biochar initiative Standards	European biochar certification test method	European biochar certification standards
	Polycyclic aromatic hydrocarbons (PAHs)/milligram per kilogramme dry weight	The United States Environmental Protection Agency 8270 (2007) using Soxhlet extraction (the United States Environmental Protection Agency 3540) and 100% toluene as the extracting solvent	6–300	DIN EN 15,527 Soxhlet-extraction with toluene and determination with GC–MS or DIN ISO 13877 Soxhlet-extraction with toluene and determination with high-performance liquid chromatography	Basic grade: <12 mg/kilogramme (indicates a minimum risk for soils and end-users) Premium grade <4 mg/kilogramme, which corresponds to the polycyclic aromatic hydrocarbon threshold outlined in the Swiss Chemical Risk Reduction Act (ChemRRV)
	Polychlorinated biphenyl/milligram per kilogramme dry weight	The United States Environmental Protection Agency 8082 (2007) or the United States Environmental Protection Agency 8275 (1996)	0.2–1	Dechlorane plus 100, HRMS or Soxhlet-extraction with toluene and determination with HRGC-HRMS based on the United States Environmental Protection Agency 8290 (2007–02)	<0.2
	Dioxins/Furans (Polychlorinated dibenzodioxins and polychlorinated dibenzofurans/nanogram per kilogramme dry weight	United States Environmental Protection Agency 8290 (2007)	17	Dechlorane plus 100, HRMS or Soxhlet-extraction with toluene and determination with HRGC-HRMS based on the United States Environmental Protection Agency 8290 (2007–02)	<20 nanograms/kilogramme International Toxicity Equivalent

Table 18 (continued)

Assessment classification	Parameter/unit	International biochar initiative test procedure	International biochar initiative Standards	European biochar certification test method	European biochar certification standards
	Heavy metals and other elements/milligram per kilogram dry weight	All according to Test Methods for the Examination of Compost and Composting (2001), while Mercury is according to the United States Environmental Protection Agency 7471 (2007)	Arsenic: 13–100, Chromium: 93–1200, Cadmium: 1.4–39, Cobalt: 34–100, Lead: 121–300, Copper: 143–6000, Molybdenum: 5–75, Mercury: 1–17, Nickel: 47–420, Zinc: 416–7400, Selenium: 2–200, Boron: Declaration needed, Chlorine: Declaration needed, Sodium: declaration needed	All metal analysis is based on microwave acid digestion with hydrofluoric acid and nitric acid and determination of the metals with ICP-MS (DIN EN ISO 17294–2) Mercury is according to DIN EN 1483 Water quality using atomic absorption spectrometry	*Basic grade:* Lead: <150, Cadmium: <1,5, Copper: <100, Nickel: <50, Mercury: <1, Zinc: <400, Chromium: <90; *Premium grade:* Lead: <120, Cadmium: <1, Copper: <100, Nickel: <30, Mercury: <1, Zinc: <400, Chromium: <80 Basic Grade following Germany's Federal Soil Protection Act (BBodSchV). Premium Grade following Switzerland's Chemical Risk Reduction Act (ChemRRV) on recycling fertilisers

Biochar must meet certain mandatory characteristics, including carbon, hydrogen, and nitrogen content, as well as hydrogen-to-carbon molar ratios. A value of 0.7 is defined as the upper limit for optimal biochar stability. Additionally, the biochar must meet certain criteria, including being free of or containing a limited range of certain heavy metals and other contaminants. *DIN* Deutsches Institut fur Normung (German Institute for Standardisation). *ISO* International organization for standardization. *ICP-MS* Inductively coupled plasma mass spectrometry. *ICP-OES* Inductively coupled plasma optical emission spectrometer. *GC–MS* Gas chromatography–mass spectrometry. *HRGC–HRMS* high-resolution gas chromatograph–high-resolution mass spectrometer

Fig. 7 Integration of biochar production and biogas production. The system illustrates potential trade-offs between climate change mitigation, carbon sequestration, biogas production enhancement and improvement, and soil stabilisation/improvement. The produced biogas can be utilised for heat, electricity, or as a vehicle fuel biomethane. The generated digestates can be used as a soil amendment fertiliser; alternatively, solid digestate-derived biochar can be used directly as a soil amendment fertiliser or indirectly to enhance and upgrade biogas. The hydrogen-assisted biogas upgradation can achieve the power to gas shifting pathway representing another integration concept
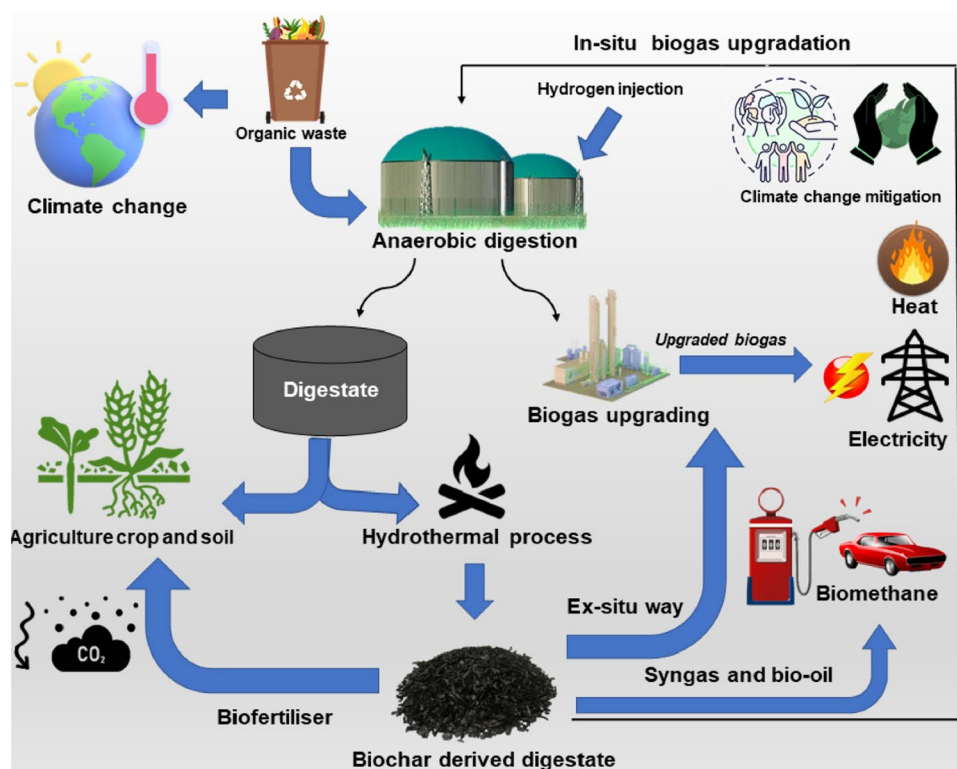
degradation through increased microbial activity, reducing ammonia and greenhouse gas emissions, increasing nutrient quality and compost maturity through organic matter stabilisation, and advancing crop productivity when compost is applied in agronomy.

- Exploiting the inherited properties of biochar, such as particle and bulk density, porosity, surface charges, water holding capacity, surface areas, and aromaticity, to buffer anaerobic digestion, mitigate anaerobic digestion inhibitors, optimise syntrophic improvements between anaerobic microbiota, and integrate other processes such as water treatment, would be an interesting area of research.
- The solubility of hydrogen gas in liquid solution and the rate of gas-to-liquid transfer are the primary barriers to hydrogen-assisted methane upgradation technology. Coupling a bio-electrochemical system with biochar is a novel area for future research in this regard.

Summary and remarks

The digestate is produced throughout the year and must be stored until the growing season of the plants. The duration of storage is determined by the region's nature, soil type, weather, crop cycle, and digestate operation protocols. The storing duration of 6–9 months is used in moderate weather. When digestate is stored in open reservoirs, methane and ammonia gases are released, reducing the fertiliser's value and causing greenhouse gas emissions. Additionally,

improper digestate management may have unfavourable consequences for public acceptance and economic viability of anaerobic digestion as a waste valorisation option, as well as for energy balance and carbon footprint. Due to several constraints, such as environmental impacts, organic matter quality, transportation costs, and nutrient availability, land spreading of digestate is insufficient to address the anaerobic digestion digestates challenge. As a result, managing massive amounts of digestate produced by anaerobic digestion became a priority.

Biochar/hydrochar from digestate can be formed via pyrolysis and/or gasification and hydrothermal carbonisation technologies as a beneficial approach to optimising anaerobic digestion byproducts. Pyrolysis and gasification processes may generate additional oil and syngas, enhancing the process's viability. Similarly, hydrothermal carbonisation can result in forming a liquid effluent along with the hydrochar. Although hydrothermal carbonisation techniques do not require the drying step required for gasification and pyrolysis, the produced hydrochar has lower carbon stability. Gasification and pyrolysis both produce biochar with higher carbon stability; however, gasification produces less biochar than pyrolysis.

As a result, combining anaerobic digestion and pyrolytic biochar is recommended. The thermal treatment modifies the chemical properties of the carbon in biochar, resulting in aromatic forms that are highly resistant to microbial degradation and stable for long periods of

(a)

(b)
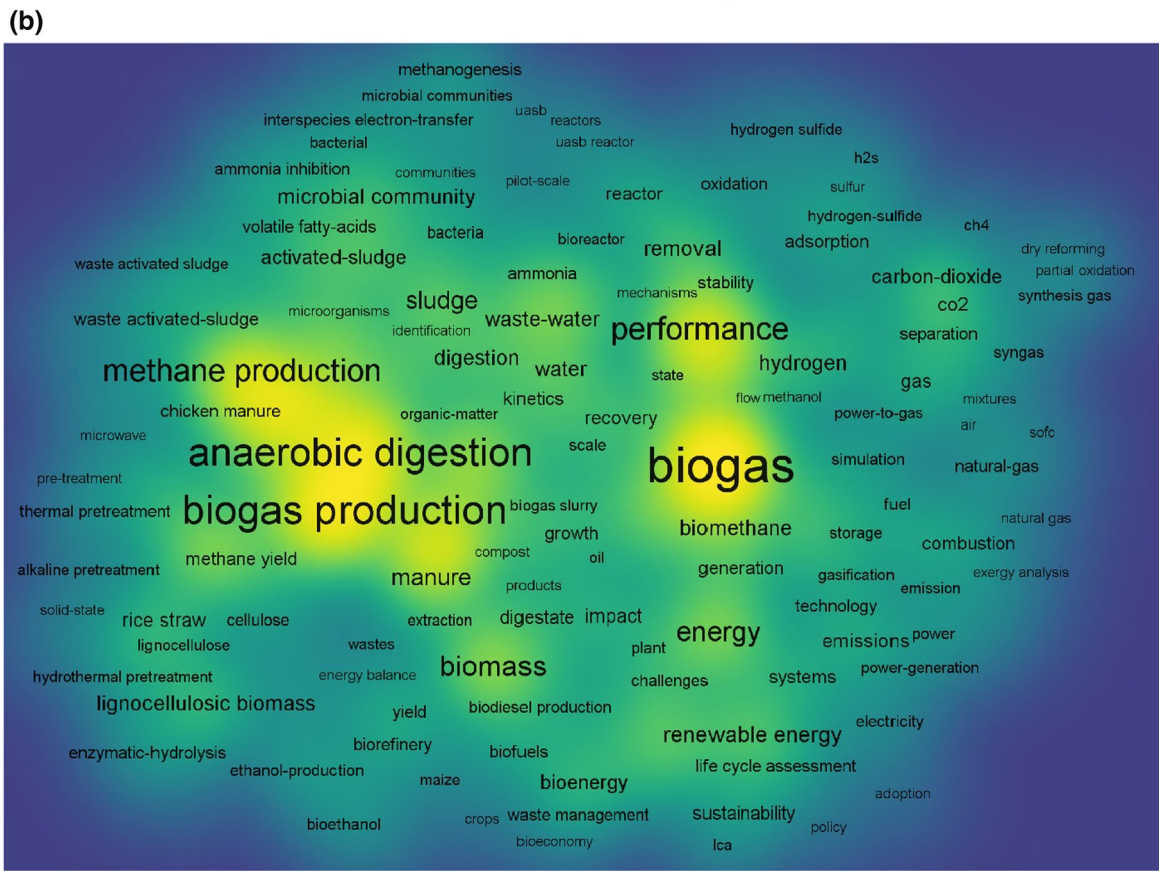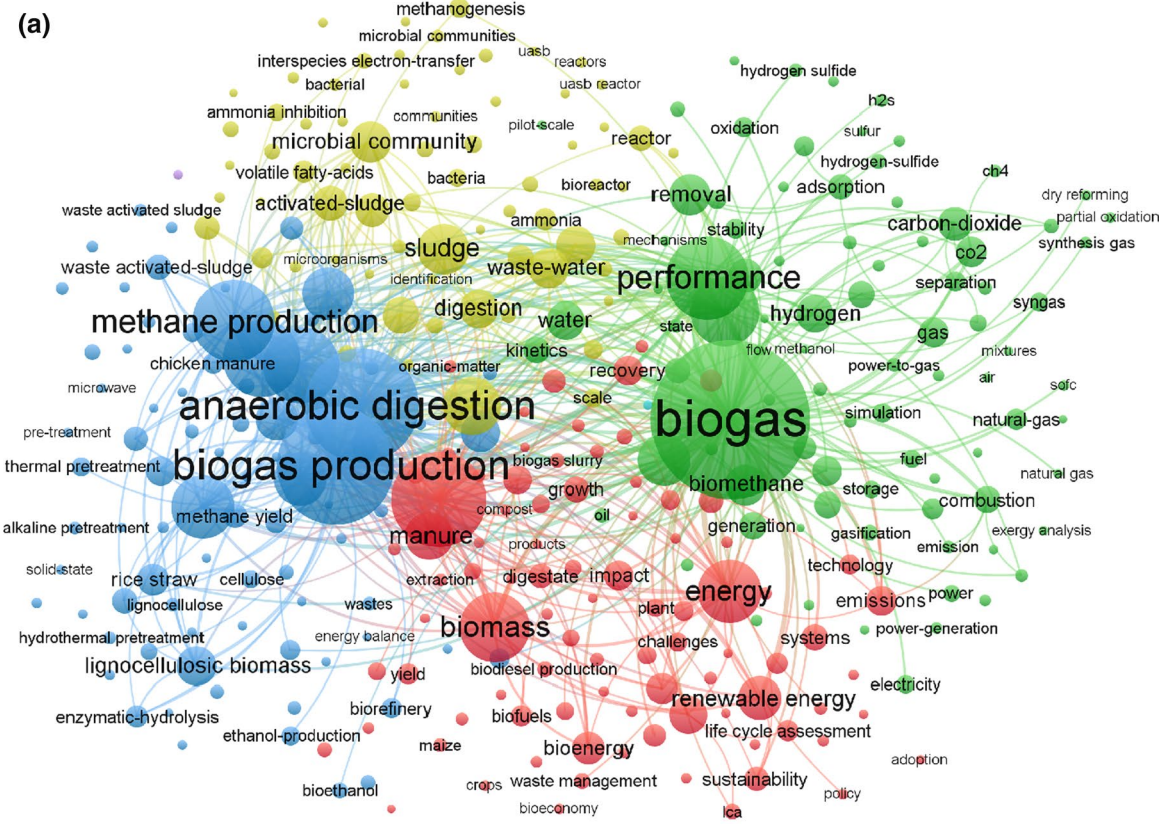
◄**Fig. 8** Bibliometric network mapping of the biogas production over the last five years, from 2018 until 2022. The data was extracted from the Web of Science. **a** Shows the network visualisation of biogas production research. **b** Shows density visualisation of biogas production research

time, hundreds of years. Thus, biochar is believed to be an effective tool for long-term carbon sequestration with the potential to significantly mitigate the effects of climate change. Divesting 1% of the net annual carbon cycled between the atmosphere and plants into biochar would significantly reduce around 10% of current anthropogenic carbon emissions. Thus, 3 gigatonnes of biochar annually produced would mitigate approximately 3 gigatonnes of carbon dioxide emissions into the atmosphere, which is consistent with the global trend for climate change mitigation. In terms of carbon sequestration, the anaerobic biochar-derived digestate would either be used directly as a soil amendment agent to replace chemical fertilisers or would be used indirectly as a biomethanation upgrading and biogas cleaning material.

Biogas bibliometric mapping

Search methodology for bibliometric mapping: the topic was biogas, and the search was performed over the last five years (2018–2202).

The bibliometric mapping provides a visual aid for researchers, whether they are experts in the field or new to the research area. The visualisation map's clusters/family trees of research terms illustrate how the entire research domain is divided or subdivided into distinct areas/research topics. The closer the clusters appear to be or are actually connected, the more direct the connection and overlap between the subtopics and research areas related to biogas production. The bigger and thicker circles in 7a represent the perceived impact of these terms or the frequency with which they are used in the literature. Additionally, the fact that they are coloured differently indicates that they are associated with distinct discrete clusters and trees that have off branching terms used in the relevant clusters.

Figures 8a, b illustrates the bibliometric mapping analysis using network and density visualisations, respectively. The bibliometric mapping analysis revealed 12,925 results from the Web of Science when using the search methodology described above. The search was conducted using a fractional count of co-occurrences or keywords that appeared 50 times in publications between 2018 and 2022. The network mapping in Fig. 8a indicates that biogas, anaerobic digestion, methane production, performance, and biomass keywords are highly prevalent. Simultaneously, those keywords with a high frequency of occurrence appear in bright yellow in Fig. 8b of the density visualisation map.

Numerous studies have been conducted in the literature on parameters associated with biogas production yields, such as pH, digestate impact, co-digestion, and activated sludge. On the other hand, the bibliometric analysis reveals a significant gap in the literature concerning membrane bioreactors, biogas for solid oxide fuel cells (SOFCs), biogas for biorefineries, microalgae cultivation for biogas, bioaugmentation, community structure, trace elements, power to gas, and techno-economic analysis of energy-efficient biogas systems.

Conclusion

Organic wastes are a global problem, with over 3.4 gigatonnes produced each year. However, just 5% of these wastes are currently being re-utilised. Devoid of effective waste management policy, these wastes would pollute our planet. This review seeks to recycle organic wastes through anaerobic digestion as a climate change mitigation policy to maintain climate warming below 2 °C. At full potential, where anaerobic digestion is digesting all readily available and unavoidable organic wastes, annual global emissions could be cut by at least 13%.

Recycling organic wastes is a win–win process, where anaerobic digestion unlocks the greatest value from organic wastes and will provide sources of renewable energy, green carbon dioxide, natural fertilisers and other valuable bioproducts, playing a multifaceted role in the circular economy's heart. Anaerobic digestion can be installed on a micro level to recycle a household's organic waste. Large-scale merchant facilities can recycle bio-wastes in cities and be a nexus of waste management and energy production, connected to local heat networks and transport fuel. Biogas can also produce heat, electricity, and fuel depending upon the geography of installations. Moreover, biogas production is continuous and does not suffer from the fluctuation of wind, solar and hydro sources, making biogas a perfect integrator to these. The biofertiliser obtained can improve both the condition and carbon capture capacity of soils and enhances biodiversity by replacing the use of synthetic fertilisers.

Biomethane quality of more than 95% is needed to utilise the generated biogas in grids, electricity, heating, or as a vehicle's biomethane fuel. To meet the subsidiaries' cascades, the biogas must be upgraded. Hydrogen-assisted, biological, biogas upgrading technology uses microorganisms as catalysts to drive carbon dioxide into methane by hydrogen utilisation. Through this concept, the hydrogen-assisted upgradation method has a great potential for transferring irregular energy (solar, wind, and raw biogas) to a more stabilised energy form (biomethane) that can store easily.

The power-to-gas shifts between the renewable technologies offer a strategy for clean energy, effective energy

storage, sustainable approach, environmentally friendly concept, and circular economy approach. This energy shift provides a long-term security aspect, where biomethane is future-proofed and would adopt tomorrow's energy needs. At anaerobic digestion's full potential, the anaerobic digestion industry could replace 33% of the demand for fossil natural gas with renewable biomethane; this proportion could be increased to 53% by integrating power-to-gas technology that converts anaerobic digestion's bio-carbon dioxide into additional biomethane. Another major benefit of this approach is that carbon dioxide is captured and recycled for new products; hence carbon capturing and sequestration are achieved, as well as the removal of carbon dioxide formed in the raw biogas without the need for exogenous energy or hazardous chemical use, required by other physical and chemical upgrading methods.

As a beneficial approach to optimise anaerobic digestion byproducts, biochar from digestate can be produced via pyrolysis technologies. Pyrolysis processes may further generate syngas and bio-oil, which could improve the viability of the full process. Pyrolytic biochar is of higher carbon stabilities up to hundreds of years; hence, biochar is believed to be a useful tool for long-term carbon sequestration with great ability for climate change crisis mitigation. The integration between the anaerobic digestion process and pyrolytic biochar is recommended as a soil amendment direct agent to replace the chemical fertilisers or as indirect biomethanation upgrading and biogas cleaning materials.

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