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Review

Biogas and Biomethane Production and Usage: Technology Development, Advantages and Challenges in Europe

Josipa Pavičić [1], Karolina Novak Mavar [2,*], Vladislav Brkić [2] and Katarina Simon [2]

[1] Pere Pirkera 38, 10361 Sesvete, Croatia; josipapavicic2003@gmail.com
[2] Faculty of Mining, Geology and Petroleum Engineering, University of Zagreb, 10000 Zagreb, Croatia; vladislav.brkic@rgn.hr (V.B.); katarina.simon@rgn.hr (K.S.)
* Correspondence: karolina.novak-mavar@rgn.hr

Abstract: In line with the low-carbon strategy, the EU is expected to be climate-neutral by 2050, which would require a significant increase in renewable energy production. Produced biogas is directly used to produce electricity and heat, or it can be upgraded to reach the "renewable natural gas", i.e., biomethane. This paper reviews the applied production technology and current state of biogas and biomethane production in Europe. Germany, UK, Italy and France are the leaders in biogas production in Europe. Biogas from AD processes is most represented in total biogas production (84%). Germany is deserving for the majority (52%) of AD biogas in the EU, while landfill gas production is well represented in the UK (43%). Biogas from sewage sludge is poorly presented by less than 5% in total biogas quantities produced in the EU. Biomethane facilities will reach a production of 32 TWh in 2020 in Europe. There are currently 18 countries producing biomethane (Germany and France with highest share). Most of the European plants use agricultural substrate (28%), while the second position refers to energy crop feedstock (25%). Sewage sludge facilities participate with 14% in the EU, mostly applied in Sweden. Membrane separation is the most used upgrading technology, applied at around 35% of biomethane plants. High energy prices today, and even higher in the future, give space for the wider acceptance of biomethane use.

Keywords: biogas; biomethane; gas cleaning and upgrading; anaerobic digestion; circular economy; emission reduction

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1. Introduction

The European Union intention to achieve a sustainable and competitive circular economy by 2050 implies structural changes of energy systems. The first step in transition towards net-zero emissions is a 55% reduction in greenhouse gases (GHG) achieved by 2030 [1]. By the Fit for 55 proposals [2], released in July 2021, the European Commission (EC) is going to require a dramatic technology revamp, through low-carbon processes engagement, comprising energy efficiency and replacing conventional energy sources with green energy sources [1,3]. However, amazing goals from today's point of view affect different sectors. Characterized by huge emissions, the transportation sector is expected to undergo significant changes. The revised Directive (EU) 2018/2001 on the promotion of the use of energy from renewable sources (REDII) [4] sets the targets for both the renewable energy share in transportation and the advanced biofuels sub-target of 14% and 3.5%, respectively, by 2030. While the proposed revision replaces the 14% renewables target with the target of 13% decreasing GHG fossil fuel intensity, it is obvious that both of the goals include a greater use of biofuels. In order to encourage waste-based biofuels production, most of the EU member states have decided to establish a double counting system in the production of biofuels from used cooking oil or animal fat. Directive restrictions cover a limitation of double counting to 1.7%. Further restrictions are focused on the elimination of biofuels produced from crops which pose a threat of significant expansion of the production area into land with high carbon stock [5].

The use of biofuels varies among the EU member states, depending on their decisions on including renewables in the transportation sector and a reduction of GHG intensity of fuels, primarily under the Directive (EU) 98/70 relating to the quality of petrol and diesel fuels [6], which was later harmonized together with RED (2009/28) [7] into Directive (EU) 2015/1513 [8]. There is, however, a different approach applied among countries. While some countries chose an overall biofuel target, others decided to separate the goals for biofuels in petrol or diesel, or they allowed themselves to rely only on fuel carbon intensity reduction targets (Figure 1). As per the National biofuels policies 2022 [8], and as can be seen in Figure 1, the majority of the countries (18 member states and the UK) set overall biofuel targets. There are 13 member states with differing goals in petrol or/and diesel, as well as six countries having both overall biofuels targets and specific targets in petrol and diesel. Advanced biofuels obligation is recognized in the biofuel policies of 20 countries. The highest biofuel targets are defined in Finland and the Netherlands, setting 19.5% and 17.5%, respectively. On the other hand, Sweden or France opted to set higher targets for reducing the GHG fuel intensity (30.5% for diesel and 7.8% for petrol in Sweden and 10% in France), without overall biofuel related goals. The UK set a 12.6% biofuel target by defining a crop cap at 3.67%, while crop-based biofuels are capped at 4.5% in Estonia. Double counting of advanced targets is not possible in Finland.

However, new strategies developed under the European Green Deal [1], such as the EU Methane Strategy [9], the Energy System Integration Strategy [10], and the From Farm to Fork strategy [11], place biogas in a central position [12]. In light of low carbon strategies, gas market retention requires zero carbon gases and decarbonisation of natural gas (methane). Decarbonized gas options refer to biogas, biomethane, synthetic natural gas, hydrogen from renewable energy, and hydrogen from the reforming or splitting of methane [13]. Biogas is a mixture of gases, primarily consisting of methane (CH_4) and carbon dioxide (CO_2), which is produced by the decomposition of organic compounds derived from different sources [14].

In the past decade, there was an increase of 90% in the biogas industry. The installed capacity globally reached 120 GW in 2019 compared to 65 GW in 2010 where more than 70% of the world biogas generation belongs to Europe [15]. According to the EBA Statistical Report 2021 [16] biogas and biomethane production amounted to 191 TWh in 2020 (18.0 bcm of energy) in Europe. This figure is intended to be doubled in 2030, and to reach over 1000 TWh in 2050, taking a share of up to 40% of the total European gas demand.

In an era of energy crisis, biomass-based bioenergy has been given special attention. Generally, a commonly used feedstock for bioenergy production is lignocellulosic biomass, as the most economical renewable source. Lignocellulosic biomass feedstock refers to municipal solid waste, forest and agricultural products, algae, etc. Lignocellulosic biomass is composed of lignin, cellulose, hemicellulose, ash, and extractives. On the other hand, sewage sludge, animal fat, and animal manure are the major constituents of non-lignocellulosic biomass, rich in protein and fatty acids with less lignin, cellulose, and hemicellulose. Although the term is usually reserved for transport fuels derived from biomass, its wider meaning refers to any fuel derived directly or indirectly from biomass and used for energy purposes that can be sold in liquid and gaseous phases. Various conversion techniques for biofuels are available or under development, reviewed by different authors [17–27].

However, this article reviews the processes deployed for biogas production, cleaning and upgrading, and use. Biogas feedstock can be grouped into agricultural waste, municipal organic waste and industrial waste. Common feedstocks include livestock manure, food-processing waste, and sewage sludge [28–30]. Substrates for biogas production differ significantly by region and commonly depend on the availability of certain raw materials in a certain area [14].

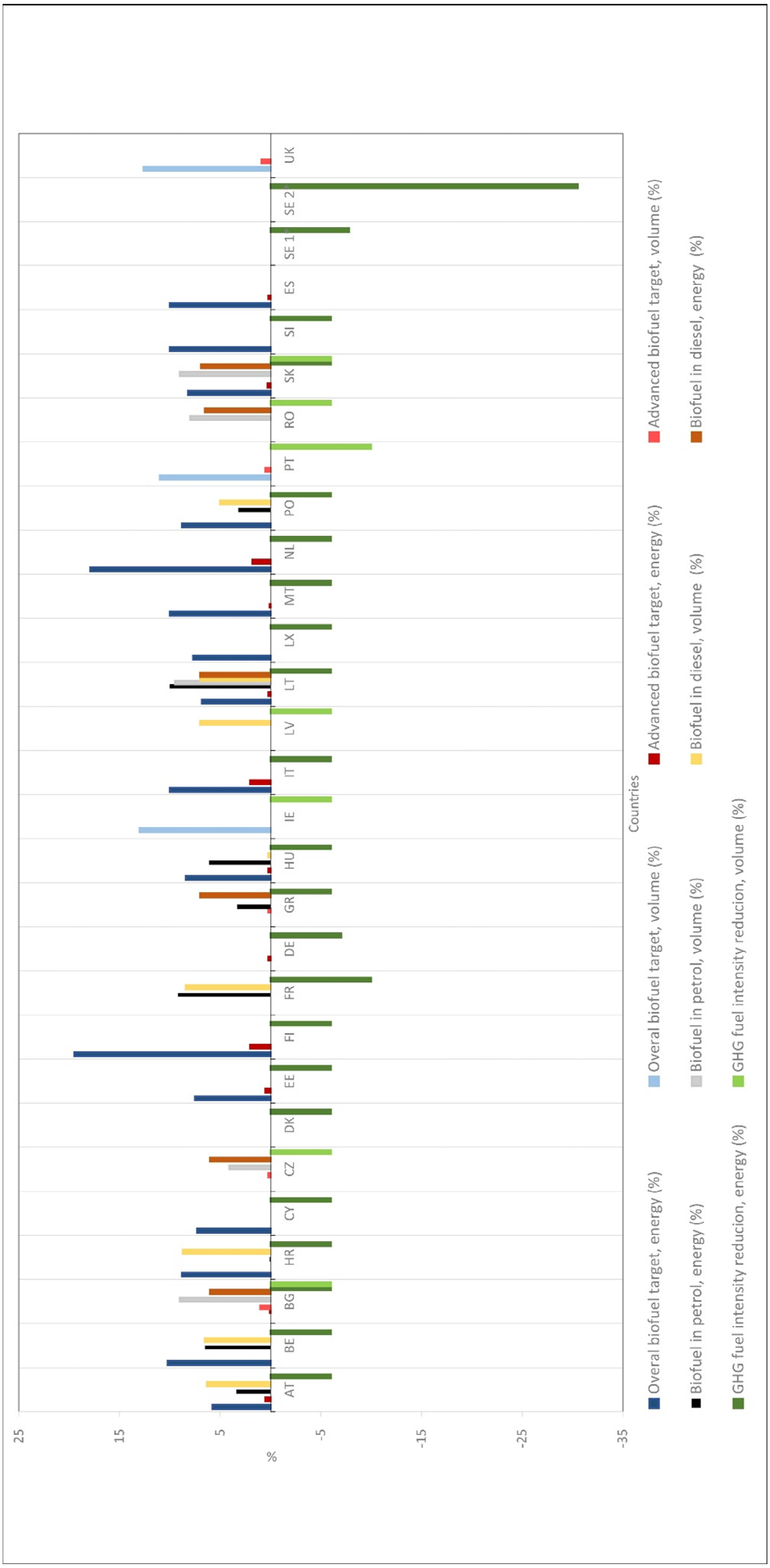

Figure 1. Biofuels targets in the EU (according to [8]).

Biogas usage is mainly connected to electricity and heat generation, and household purposes (cooking and heating). After upgrading to biomethane, it can be injected into a gas grid or used as transport fuel, which is expected to be its main future purpose. Installations for heat and power generation with electrical capacity are in the range from tens of kWe to a few MWe. Different energy applications of biogas are shown in Figure 2. After advanced upgrading, biomethane can be injected into a natural gas pipeline grid, used as a transportation fuel or in fuel cells. Compression and liquefaction are needed to convert biogas into compressed natural gas (bio-CNG) and liquefied biogas (LBG), while catalytic reforming is a commonly applied chemical method to obtain syngas. By Fischer–Tropsch synthesis (FTS) or fermentation process, syngas can be transformed into alcohols, such as methanol or ethanol [15,31–34].

About 90% of globally produced biomethane is generated by the anaerobic digestion process, followed by different upgrading treatments [31]. Production alternatives include thermal and hydrothermal gasification. Engagement of pretreatment results in a wider range of suitable input raw materials and process efficiency improvement [35,36].

Upgrading to biomethane or renewable natural gas (RNG) is being adopted in developed countries, especially in North America and Europe. Such projects are often developed by oil and gas companies in the USA for gaining carbon credits. As a vehicle fuel alternative, biogas has already been applied in the European Union (Sweden and Germany) and the USA, either as pure methane (CBG100) or in a mixture with natural gas (CBG10 and CBG50) [15,37,38].

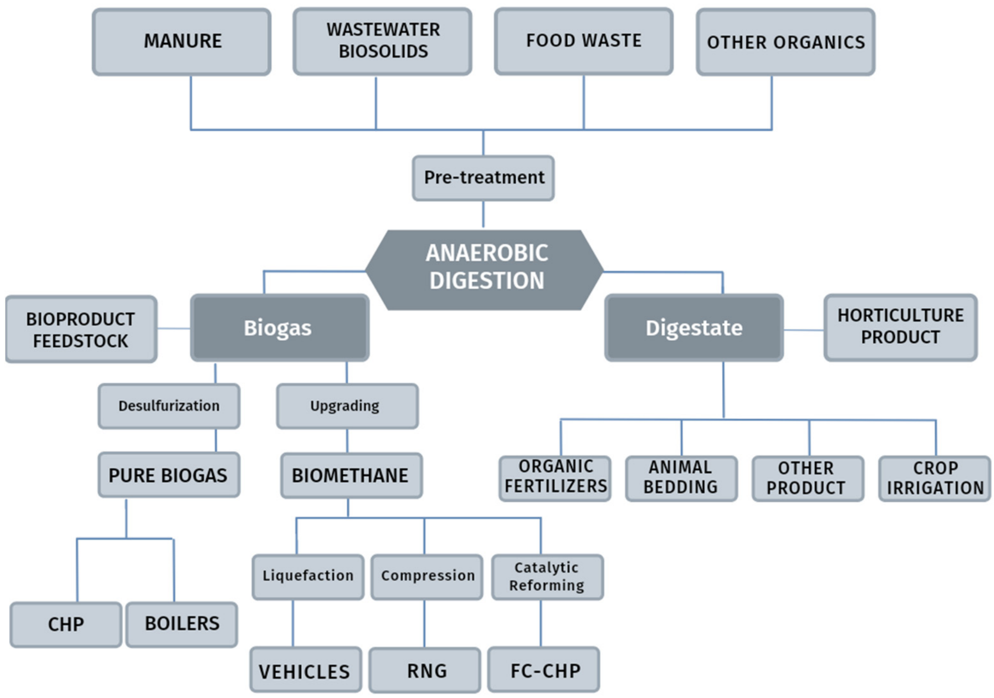

Figure 2. Possibilities of using biogas.

The aim of this paper is to give a representative overview of the overall process of biogas and biomethane production, from feedstock used to the final product in order to get a foundation for further research. In line with that, the paper gives biogas composition, analyses the substrates options that results in satisfactory yield and available technologies for biogas and biomethane production. The analysis is made based on published and publicly available data, with the focus on the most often-used anaerobic digestion process and pretreatment methods. Conventional upgrading techniques are elaborated, but also some modern, cost effective and environmentally friendly solutions are shown. Biogas and biomethane temporary usage and further streams are given through a literature review on papers published in the last five years.

Another target is to analyse the role of biogas in achieving a climate-neutral Europe, through determination of the current position of biogas in the energy mix and defining its perspective in light of new strategies and geopolitical developments. For that purpose, the EU legal requirements in terms of biogas usage and emission reduction are analysed. Based on data published by different organisations, analyses on biogas presence in Europe, along with leading production countries and distribution of the installed biomethane facilities per both feedstock and upgrading techniques, are elaborated.

2. Methods

A literature review was used as a methodology for conducting research. The intention was to make a comprehensive review of technology applied for biogas production and usage. Therefore, the authors have tried to collect data on the available processes, not only those which are commonly in use, but also those that are currently being under research and development. Additionally, a systematic literature review was initiated to identify relevant data related to biogas cleaning and upgrading techniques, and possible pretreatments used to improve process efficiency. A separate focus was on detecting the biogas current energetic position, as well as its future potential in the EU, keeping in mind the required transition to a low carbon economy.

Turbulent developments of climate-policies, technology progress and a recent upturn in biogas facilities are the reason that we have mostly, but not exclusively, focused on in recent publications. Although most of the considered literature was published in the period from 2015 to 2020, some of the earlier published papers were found convenient due to their information regarding the general technology characteristics.

This review paper covers data obtained from academic literature and professional reports issued by different agencies and organisations. Academic literature covers peer-reviewed papers, books, theses, abstracts and technical reports containing exact figures and have served as an accurate basis in our speculation on technology prospects. The statistics related to the status of biomethane facilities were mostly adopted from reports published by different professional associations and agencies. The figures are additionally analysed, rearranged and compiled in order to analyse actual developments.

Construction of this article ensures a clear review on technology development, divided into separate process phases, i.e., pretreatment, production, purification and application of biogas. Each chapter concludes with an analysis of future technology trends.

3. Biogas Production

3.1. Biogas Composition

Biogas streams mainly consist of methane (CH_4, up to 75%) and carbon dioxide (CO_2, up to 50%), while hydrogen (H_2), nitrogen (N_2), water vapour ($H_2O_{(g)}$) and hydrogen sulphide (H_2S) may be present in a smaller proportion. Biogas composition and its properties depend on the substrate type (source of biomass entering the process), the type of facility and the process conditions. Table 1 compares the composition of raw biogas and natural gas. Individual volume shares differ to a lesser extent depending on the literature source.

Table 1. Biogas vs. natural gas composition [14,39–41].

Compound	Formula	Volume share (%)			
		Biogas			Natural gas
		Korbag et al. [14]	Persson et al. [39]	Moya et al. [40]	IEA Bioenergy [41]
Methane	CH_4	50–75	53–70	55–70	83–98
Carbon dioxide	CO_2	25–50	30–47	30–45	0–1.4

Water	H_2O	5–10	/	/–5	
Oxygen	O_2	<2	0	0–3	
Nitrogen	N_2	<10	0.2	<15	0.6–2.7
Ammonia	NH_3	<1	<1 (<100 ppm)	<1 (0–100 ppm)	/
Hydrogen	H_2	<1	0	/	/
Hydrogen sulphide	H_2S	<3	<1 (0–10,000 ppm)	<1 (0–10,000 ppm)	/
Ethane					<11%
Propane					<3%

In addition to the listed substances, traces of iron, nickel, cobalt, selenium, molybdenum and tungsten can be present [42]. The chemical composition of biogas will also determine the process that needs to be applied to optimize its usage properties. The presence of CO_2 and N_2 negatively affect the gas calorific value, while CO_2, H_2S and water vapour form corrosive conditions, which cause equipment damage [14].

3.2. The Biogas Production: From Substrate to Biomethane

The chemical composition and biodegradability of the used substrates affect the biogas composition. Generally, substrates rich in lipids and proteins have a greater potential of methane yield compared to those rich in carbohydrates, but high lipid content leads to the anaerobic digestive system failure due to long-chain fatty acid generation. On the other hand, substrates rich in carbohydrates can negatively affect the C/N ratio, resulting in nutrient restriction and quick acidification [30]. Formerly, only one input substrate entered the process, but recent practice prefers a combination of different substrates: the so-called co-digestion. Co-digestion has led to a higher gas production per ton of substrate [43].

Livestock manure is commonly considered a cost-effective and viable feedstock for biogas production, but differences in animal digestive systems, nutrition or manure storage mechanisms result in different and most often lower biomethane production. Although an abundance of lignocellulosic biomass (crop residues, forest residues, energy crops, etc.) could serve as a good basis for sustainable energy production, lignin in lignocellulosic biomass encapsulates hemicellulose and cellulose into a hydrophobic structure of Lignin-Carbohydrate Complexes (LCC), requiring the application of suitable pretreatments. Regarding industrial waste, waste from the food industry has been commonly used in bioreactors due to its chemical properties and biodegradability. Figure 3 represents the selected substrate types used in anaerobic digestion that pose good biomethane yield in mL/g volatile solids (VS). Among a large number of possible substrates, only those with an average biomethane yield over 250 mL/g VS are shown. Livestock manures generally provide a lower methane yield (around 200 mL CH_4/g VS) due to higher ammonia inhibiting compounds (sheep manure and goat manure substrates show the best results). On the other hand, food by-products pose a significant biomethane yield (over 500 mL/g VS). Food by-products rich in lipid content (kitchen waste) have a higher methane yield. To satisfy the nutritional requirements of microorganisms, a low C/N ratio characterized for livestock manure is combined with carbon rich feedstock, achieving satisfactory results in biomethane yields [44].

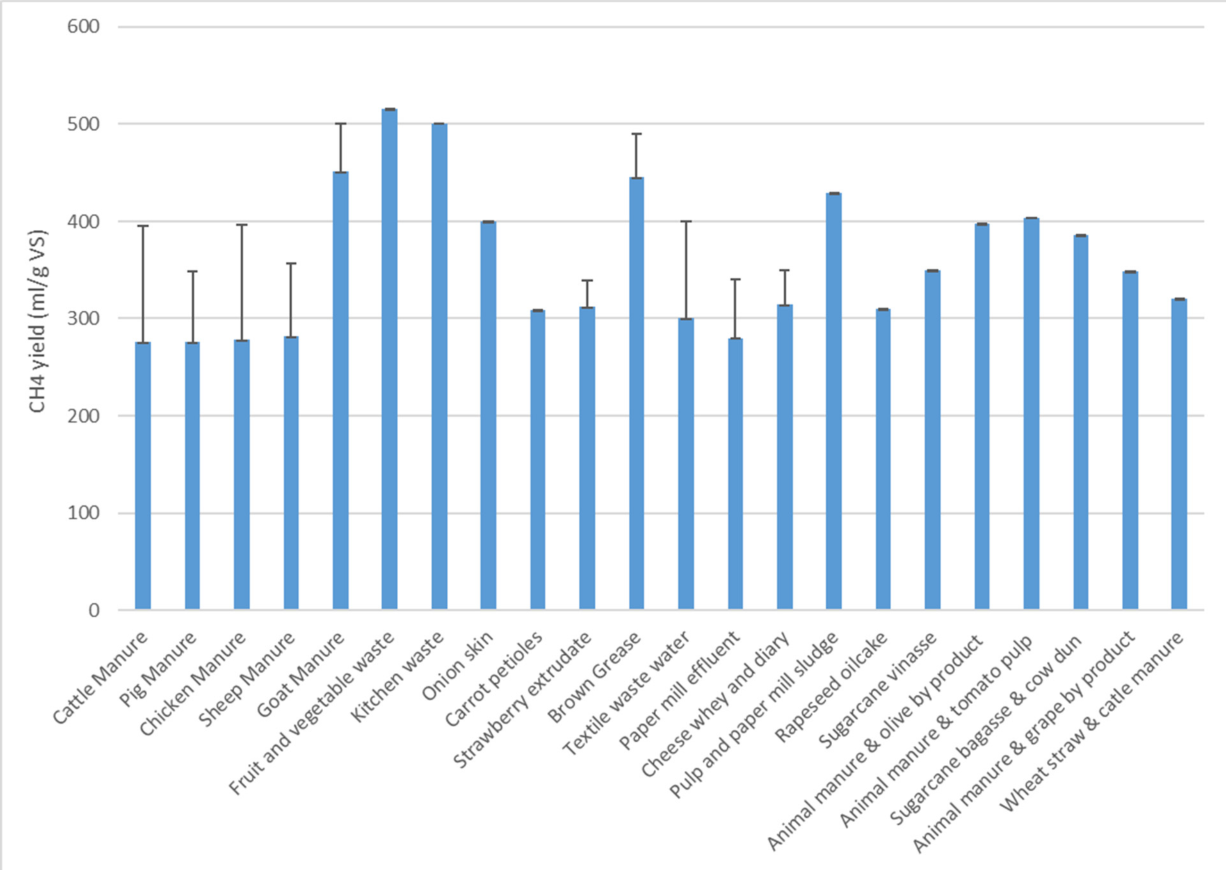

Figure 3. Biogas yield from different substrates with biomethane yield above 200 mL/g VS (according to [30]).

Recently, production plants have been more oriented towards the use of municipal and industrial waste. However, manure and sludge from wastewater are the most common input (80%) [14]. Although AD is a promising technique for waste sludge management [4], complex microstructure components make sewage sludge difficult to hydrolyze and digest. Components such as extracellular polymeric substances have low volatile solid degradation (30–50%) even with a long retention time, which leads to low methane yield. Biomethane productivity is enhanced by different pretreatment options and by co-digestion with organic wastes [45–47].

Song et al. [48] determined the optimal mixing ratio of 2:1:1 for sewage sludge, food waste, and livestock manure by applying the biochemical methane potential (BMP) test, Design Expert software and continuous reactor operation.

Process input varies significantly by region, being imitated by those materials that are locally available due to transport costs minimization [49,50].

3.3. Using Technologies in Biogas and Biomethane Production

Although AD with upgrading treatments is a commonly used technology in biomethane yielding, which means that 90% of the produced biomethane comes from AD, other alternatives are commercially available, and some emerging technologies can be found. The International Energy Agency (IEA) divides biogas based on the feedstock and treatment type into sewage sludge gas, landfill gas and other AD biogas (methanization of non-hazardous waste or raw plant matter) [31].

3.3.1. Anaerobic Digestion

Anaerobic digestion is the biochemical process of decomposition of organic compounds by microbe bacteria under anaerobic conditions. The process itself is actually similar to some natural processes, such as peat formation, or the process in the ruminant digestive system. Methane fermentation requires diverse associations of bacteria, i.e., aceticlastic and hydrogenotrophic methanogens, syntrophic acetogens, fermentative bacteria and homoacetogen [15,51].

Biogas production can be accomplished via two main fermentation processes. In the dry process, the solid concentration in the fermenter is lower than 10%, and in the wet process, it ranges from 15 to 35% [15,52]. However, different factors affect the AD process, such as temperature, pH value, carbon to nitrogen ratio (C/N) and substrate particle size. It can be expected that smaller particles undergo faster decomposition, resulting in faster biogas generation. The process pH ranges from 5.5 to 8.5, with optimal results obtained at higher values. As per Sahota et al. [53], the optimal C/N ratio is in the range from 20/1 up to 30/1 [54,55].

An AD environment can be generally divided into psychrophilic (low temperature), mesophilic (moderate temperature), and thermophilic (warm). Each regimen is suitable for a specific type of microorganism. The psychrophilic process takes place at temperatures less than 30 °C, the mesophilic process occurs in the temperature range of 30 to 42 °C, while the temperature ranges from 43 to 55 °C is needed for the thermophilic process. However, too low or too high temperatures are not suitable due to the fact that they slow down the process [14,42,55].

Mesophilic and thermophilic regimes are more commonly used, and the temperature in these processes is maintained by installed heat exchangers. Some of the advantages of thermophilic biogas production are a higher rate of methanogenic bacteria, shorter digestion time, higher efficiency and better substrate utilization. In general, higher temperatures result in higher biogas yield and shorter retention time. However, higher energy needs require optimal conditions to be defined. Furthermore, the risk of ammonia inhibition and irreversible acidification caused by volatile fatty acid accumulation are important reasons for closer supervision of thermophilic digesters [51,56]. While conventional thermophilic anaerobic digestion optimization focuses primarily on the development of robust microbiome and co-digestion, new approaches consider integration with microbial electrochemical systems and the use of conductive additives [56].

A comprehensive review of reactors for biogas production is given by Banerjee et al. [55]. The authors stated that one-stage systems are usually installed in industry due to their performance simplicity, although two-stage systems are less energy demanding.

As seen in Figure 4, the biogas production procedure includes four important phases: hydrolysis, acidogenesis, acetogenesis, and methanogenesis [57,58]. Principal reactions which occurred in each phase are shown in Table 2. In the first phase, i.e., hydrolysis, complex organic components, such as fats, proteins and carbohydrates, are broken down into simpler molecules, i.e., amino acids, fatty acids and sugars. Equation (1) represents the hydrolysis of cellulose ($C_6H_{10}O_5$) via the addition of water (H_2O) to form glucose ($C_6H_{12}O_6$) and H_2. Chemical reactions take place inside the fermenter (digester) under bacterial activities. Acidogenesis, or the second phase, considers decomposition of chemical compounds formed by hydrolysis into methanogenic compounds, i.e., hydrogen, alcohols, carbon dioxide, carbon acids and ammonia. This is about the fermentation stage, presented by the Equations (2)–(4). Acetogenesis and methanogenesis often take place simultaneously. Carbon dioxide, hydrogen and acetic acid are formed during acetogenesis, as presented by Equations (5)–(7). However, a clear boundary between acidogenesis and acetogenesis is often impossible since both phases result in the generation of H_2 and CH_3COO—which are methanogenic bacteria substrates. Methanogenesis phase results in CH_4 and CO_2 generation. The conversion of CH_3COOH into CH_4 and CO_2 is shown by Equation (8). CO_2 is further reduced to CH_4 through H_2 (Equation (9)). The production of CH_4 by decarboxylation of CH_3CH_2OH is presented by Equation (10) [58].

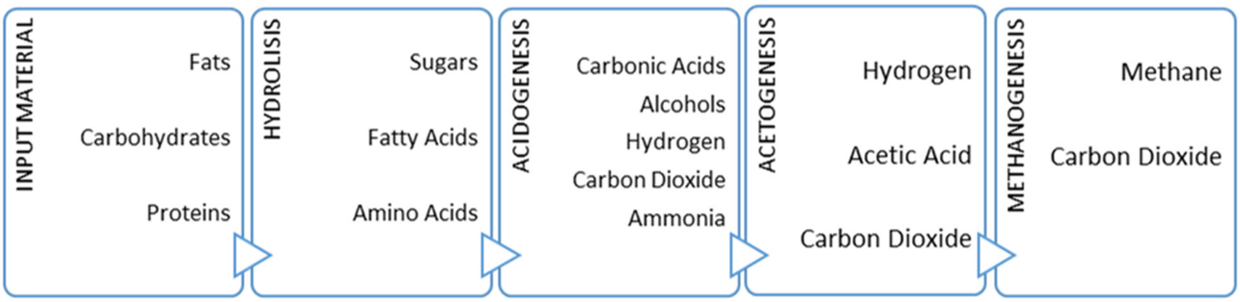

Figure 4. Anaerobic digestion process phases (modified to [51]).

Table 2. Principal reactions in the AD process (according to [58]).

AD process stage	Chemical reactions	
Hydrolysis	$(C_6H_{10}O_5)n + n\ H_2O \rightarrow n\ C_6H_{12}O_6 + n\ H_2$	(1)
Acidogenesis	$C_6H_{12}O_6 \leftrightarrow 2\ CH_3CH_2OH + 2\ CO_2$	(2)
	$C_6H_{12}O_6 + 2\ H_2 \leftrightarrow 2\ CH_3CH_2COOH + 2\ H_2O$	(3)
	$C_6H_{12}O_6 \rightarrow 3\ CH_3COOH$	(4)
Acetogenesis	$CH_3CH_2COO^- + 3\ H_2O \leftrightarrow CH_3COO^- + H^+HCO_3^- + 3\ H_2$	(5)
	$C_6H_{12}O_6 + 2\ H_2O \leftrightarrow 2\ CH_3COOH + 2\ CO_2 + 4\ H_2$	(6)
	$CH_3CH_2OH + 2\ H_2O \leftrightarrow CH_3COO^- + 3\ H_2 + H^+$	(7)
Methanogenesis	$CH_3COOH \rightarrow CH_4 + CO_2$	(8)
	$CO_2 + 4\ H_2 \rightarrow CH_4 + 2\ H_2O$	(9)
	$2\ CH_3CH_2OH + CO_2 \rightarrow CH_4 + 2\ CH_3COOH.$	(10)

Mao et al. [41,59] investigated the latest trends in AD and anaerobic membrane bioreactors (AnMBRs). Even though the intention of breaking down the comprehensive AD process into two groups, i.e., acetogenesis–methanation and hydrolysis–acidogenesis separated reactors, enhances the rate of the conversion process, this would result in higher costs [60].

3.3.2. Thermal and Hydrothermal Gasification

Thermal gasification is passing through its early commercialization phase. Such a process uses dry woody and lignocellulose biomass. Under controlled oxygen and steam, thermal breakdown occurs, resulting in syngas and biochar. Conversion into biomethane is done in a catalytic reaction or through biological processes [36,43,61]. Another option is hydrothermal gasification. It uses dry or wet raw material, which is then heated and compressed to supercritical conditions, favourable for methane rich biogas generation. Hydrothermal gasification demonstration projects in France and the Netherlands represent the forerunners of industrial application, which can be expected in forthcoming years [35,36].

3.3.3. Process Pretreatments

Pretreatment is used to overcome the obstruction of digestion. Complex organic matter needs to be solubilized and hydrolyzed into simple components, i.e., to be prepared for microbial activities. Cellulose and hemicellulose are polysaccharides that can be hydrolyzed to simple sugars. Lignin serves as a support to the cell structure and thus impedes the proneness to microbial attack during the hydrolysis process. Pretreatment processes break the lignin layer, making the biomass prone to digestion. The other role is in decreasing the crystallinity of cellulose and increasing porosity [62]. There are different advanced pretreatments, which enable a broader range of input raw materials, and improve the AD process efficiency by faster digestion and yield increasing. A decrease in the retention period leads to economically reasonable small-scale reactors.

Various in situ and ex situ pretreatment techniques are used in order to achieve better results [15,44,62–67]. Pretreatment methods can be grouped into physical, chemical, physico-chemical and biological methods. (Table 3).

Table 3. Types of pretreatments most used to improve biogas production [62–66].

Pretreatment type	Method	Mode of action
Physical	Milling Extrusion Cavitation Microwave Irradiation	Biomass is altered by cells breaking when physical force is used.
Physico-chemical	Liquid Hot Water Steam Explosion Ammonia Fiber Expansion CO_2 Explosion	A combination of mechanical forces and chemical effects applied to biomass.
Chemical	Alkaline Hydrolysis Acid Hydrolysis Ionic Liquids Organosolv Wet Oxidation	Use of organic or inorganic compounds to disrupt the structural recalcitrance of lignocellulosic biomass.
Biological		Enzyme-producing fungi are used to alter biomass.

Physical pretreatment methods reduce the particle size resulting in an increase in surface area and a decrease in polymerization degree and crystallinity. Increased surface area improves accessibility to microbial and enzymatic degradation activities. The main disadvantages of physical pretreatments are in their inability to remove the lignin content of lignocellulosic biomass and their high energy request [58,62–68]. Chemical pretreatment of biomass considers different chemical compounds to be applied in order to accomplish a disruption of biomass structure through interaction with intra and inter polymer bonds. The lignocellulosic biomass structure disruption results in cellulose crystallinity reduction, depolymerisation and degradation of cellulose and lignin breakdown. Alkali Hydrolysis is based on the solubilization of lignin in the alkali solution (NaOH, $Ca(OH)_2$, NH_3). Acid Hydrolysis takes advantage of the sensitivity of glucosidic bonds between hemicellulose and cellulose to acid (H_2SO_4, HCl, CH_3COOH, HNO_3). The organosolv process uses organic solvents for breaking down the bonds between lignin and hemicellulose. Wet Oxidation considers applying oxygen in the temperature range of 140–200 °C. It enhances contact between molecular oxygen and organic matter and thus achieves the complete degradation of organic compounds into CO_2, H_2O and simpler, more oxidized organic compounds into low molecular weight carboxylic acids [58,62–68]. Physico-chemical methods represent both mechanical forces and chemical effects applied to biomass. In the Steam Explosion, bioreactors are saturated with steam at 160–200 °C and 0.69–4.83 MPa. Steam condensation and its penetration into biomass causes sudden depressurization, breaking of glyosidic hemicellulose bonds and solubilization. Liquid Hot Water is similar to Steam Explosion, using water at elevated temperatures (170–230 °C) and pressure (up to 5 MPa). The pressure application is due to maintaining the liquid phase. The process hydrolyses hemicellulose by liberating its acetyl groups and removes lignin. Ammonia Fiber Explosion uses NH_3 at relatively high temperatures (90–100 °C) and moderate pressure for approximately 30 min, before its rapid release. The rapid release of pressure disrupts the fibrous structure of biomass and decrystallizes the cellulose, hydrolyses hemicellulose, removes and depolymerises lignin, and increases the size and number of micropores and improves the accessibility of enzyme. The CO_2 Explosion uses supercritical CO_2 as a "greener" alternative compared to ammonia [69]. CO_2 acts as a solvent in the pretreated biomass, transforming it into glucose through the enzymatic hydrolysis of cellulose. The pretreatment process is not satisfactory to biomass with no moisture content, since CO_2 dissolved in water forms carbonic acid, which catalyses the hydrolysis of hemicellulose. Pressurized gas release is responsible for breaking the compact matrix structure of biomass [70]. Low temperatures are required, and no flammable or corrosive products generation are the main process advantages [58,62–68]. Biological pretreatments use microorganisms, i.e., fungi and bacteria (brown-, white- and soft-rot fungi, actinomycetes and bacteria) to degrade lignin and hemicellulose, while leaving the cellulose undamaged. Enzymes secreted by the fungi are responsible for lignin degradation. The main advantages of biological pretreatments refer to the absence of toxic substance generation and the low amount of required energy. However, the long duration of the process, the need for vigilant control of conditions and large space occupancy are the main disadvantages [58,62–68].

Thermal, thermal-alkaline, alkaline and electrochemical pretreatments are found to be the most effective for sewage sludge solubilizing. However, other pretreatment methods (such as ultrasonication, microwave and high-pressure homogenization) are being considered. When it comes to food waste, a hybrid biological-physiochemical treatment is suggested. Since the manure consists of lignocellulose fibres; its pretreatment is similar to that for energy crops/plant residues [71–73].

By applying different chemical additives, such as NaOH, $Ca(OH)_2$, NH_4OH, or H_3PO_4, process stability can be improved and up to 40% higher methane yield can be achieved [15]. According to Abraham et al. [74], enhanced biogas yield of 1200% can be achieved if ionic liquid is used in pretreatment process.

Rodriguez et al. [75] investigated mechanical, microwave, thermal, chemical and biological pretreatment methods applicable for grass structure, while one of the latest studies [76] includes convenient pretreatments for improving biogas production of macroalgae. Fucus vesiculosus and Fucus serratus, as the dominant species of brown seaweed growing in the Baltic Sea, were used there. The research included mechanical, microwave (600 W, 2 min), ultrasonic (110 V, 15 min), and microwave combined with ultrasonic (600 W, 2 min; 110 V, 15 min) treatment applied to the seaweed in order to track methane yields from AD. The results showed that the ultrasonic, ultrasonic combined with microwave, and microwave pretreatments increase cumulative methane yields of 167%, 185%, and 156%, respectively. The combined pretreatment showed a maximum methane yield of 260 mL/g of volatile solids after 20 days of digestion. The ultrasonic combined with microwave pretreatment showed a significant improvement in methane yield when compared with the mechanical pretreatment.

3.4. Biogas Puification and Upgrading

More review reports on biogas purification and upgrading were issued in the last decade, covering a comparison of different commercial upgrading techniques by their efficiency and barriers [77–79]. In a detailed review, Sun et al. [80] elaborated biogas purity, CH_4 recovery and loss, upgrading efficiency, investment and operating costs.

However, conventional biogas upgrading is based on physical/chemical technologies, which result in high CH_4 concentration (88–98%) and high removal efficiency for H_2S, halocarbons, and siloxanes (min. 99%). Unfortunately, typical high energy and chemical demands affect environmental and economic sustainability [81]. Conventional biogas purification and upgrading can be divided into sorption and separation processes; they are shown in Tables 4 and 5.

Table 4. Biogas purification methods (modified to Adnan et al. [82]).

Method		Working principle
Absorption	Water scrubbing	Different solubilities of H_2S and CO_2.
Adsorption	In situ chemical precipitation (iron salts)	Chemicals used dissolves sulphides into either insoluble metallic sulphide compounds or elemental S.
	Metal oxides Al, Fe, Mn, Co, Cu, Zn oxides)	H_2S adsorbs metal oxides by the sulphur binding as metal sulphide; Efficient in mercaptans removal.
	Activated carbon (impregnated, virgine)	Catalyze H_2S oxidation into elemental S.
Biological		Microorganisams are used to convert H_2S into S.
Membranes		Selective selectivity characteristics of CH_4 and H_2S.
In-situ microaeration		Adding O_2 or air directly in digester.

Table 5. Biogas upgrading methods.

Method		Working principle
Absorption	Water scrubbing	Different solubilities of H_2S and CO_2.
	Physical scrubbing	
	Chemical scrubbing (amines)	Chemical reactions of amine mixture with CO_2.
Adsorption	Pressure swing adsorption (PSA) Vacuum swing adsorption (VSA)	Pressure changes in different process stages. VSA adsorption occur near atmospheric pressure.
	Temperature swing adsorption	Temperature changes in different process stages.
Membranes (gas–gas separation, gas-liquid separation)		Selective selectivity characteristics of CH_4 and H_2S.
Cryogenic separation		Different boiling temperatures of CO_2 and CH_4.

Physical scrubbing means that CO_2 is dissolved in a liquid under pressure (various commercially available physical solvents), while chemical scrubbing means that CO_2 reacts with chemical solvents (amines) to separate it from CH_4. The operation of scrubbing begins with removing hydrogen sulphide and other impurities. Water scrubbing is widely used for biogas upgrading. Water is used as a solvent for CO_2 because within it, there is a higher solubility of CO_2 than solubility of CH_4 [83,84].

Depending on the type of amine solution used, different reactions may occur. However, the most commonly used amine mixture is aMDEA (methyl diethanolamine + piperazine), as CO_2 absorption rate is quite high compared to other available amines.

Lee et al. [85] made a performance comparison of the removal efficiency of CO_2 and H_2S from biogas by blending amine absorbents. The research, which included twelve different absorbents, showed that CO_2 absorption and regeneration rates were excellent in MDEA/piperazine, while H_2S absorption and regeneration rates were excellent in MDEA/2-amino-2-methyl-1-propanol. MDEA-based blending absorbent showed better absorption and regeneration performance than MDEA, and MDEA/PZ showed good performance for CO_2 but very poor performance for H_2S.

Adsorption is a mass transfer process, a phenomenon of sorption of gases or solutes by solid or liquid surface. It is often preferred in gas purification due to its simplicity, relatively low-cost, flexibility, high energy-efficiency (>85%), high selectivity and regeneration possibility [86–88]. Adsorption technologies and material commonly used are shown in Tables 4 and 5. Among such processes, pressure swing adsorption appears to be the most frequent. Separation of CO_2 and CH_4 is obtained based on the difference in their attraction to an adsorbent surface. A pressure increase results in adsorbed gas. Activated carbon is commonly applied due to low costs, high specific area and porous space, and H_2S and CO_2 selectivity [87,89]. Furthermore, carbon can be produced from biomass residues and modified with metal oxides representing additional benefit. Sawalha et al. [90]

published the study on removing H_2S from biogas by adsorption using synthesized activated carbon prepared of locally available biomass. The best results in removal efficiency were achieved with eucalyptus (690 mg H_2S/g adsorbent), almond shells (230 mg H_2S/g adsorbent) and coffee grains (22 mg H_2S/g adsorbent).

Besides carbon, zeolites are also preferable due to their high selectivity, regenerability, thermal resistance and separation of H_2S through chemisorption at very low partial pressures and physisorption at low to moderate partial pressures [91,92].

Furthermore, research studies on metal-organic-frameworks (MOFs) have shown their great adsorption and selection properties toward H_2S and CO_2 [93,94]. Nowadays, the MOFs are subject to various research on their application in different gas separation processes. Taddei & Petit [95] made a review on technical information on MOF design, synthesis, characterisation and sorption testing. According to Alonso et al. [96], biogas purification and CH_4 storage would become a new motivation for the development of nanomaterials as sorbents. Their review on the current use of novel nanomaterials as adsorbents for CO_2 and CH_4 shows that materials modified with amine or metals are most perspective. The Fe_3O_4-graphene and the MOF-117 based nanoparticles show the greatest CO_2 sorption capacities, due to their high thermal stability and high porosity. IRMOF-6, MOF-177 and MOF-5 showed the highest adsorption capacities for CH_4.

Separation methods consider different membrane technologies and cryogenic separation. Membrane separation is based on a permeation process. This technology is well known and used in natural gas processing [97,98]. Comparison analysis made for organic, inorganic and hybrid materials, shows advantages of inorganic materials in terms of temperature stability, permeation rates and selectivity [99].

Cryogenic (low-temperature) purification of biogas uses very low temperature (−70 °C) and high pressure (approx. 40 bar) to remove the various undesirable components, including CO_2, H_2S, water vapour and siloxane. Process principles are based on the differences in the constituents' boiling temperature. Different gases condense at different temperature-pressure domains. Usually, the main gas stream is cooled down in a multistage refrigeration sequence, below a certain cryogenic temperature according to operating pressure. Acid components are concentrated in the liquid effluent at the bottom of distillation column. Numerous and expensive process equipment, such as compressors, turbines, heat exchangers and coolers, represent the main disadvantages of this method [100–102].

Modern developments in biogas purification and upgrading techniques have been reviewed by different authors [14,81,103]. Rodero et al. [81] stated that technologies which use aerobic and anoxic biotrickling filters, algal-bacterial photobioreactors and digesters under microaerobic conditions, successfully remove H_2S (>99%). They also made a comparison of various modern technologies for the biogas purification of H_2S, halocarbons, and siloxanes. In the same article the authors stated that biological CO_2 removal processes, including microalgae-based CO_2 fixation, H_2-assisted litoautotrophic CO_2 bioconversion to CH_4, enzymatic CO_2 dissolution or fermentative CO_2 reduction, have recently appeared as cost effective and environmentally friendly solutions, resulting in CO_2 removal of 80–100% and CH_4 purities of 88–100%.

3.5. Biogas Usage

The very first examples of biogas usage date back to the Assyrian empire, about 3000 years ago. The biogas, obtained by decomposing organic material, had been initially used for water heating. Later, in the 17th century, more intensive research of biogas and its properties began. The first modern digestion plant was recorded in India in the second half of the 19th century [104,105]. A literature review on papers published over the last five years with the topic of biogas usage possibilities and the related issues are shown in Table 6.

Table 6. Biogas usage.

Biogas usage	Topics elaborated in recent publications on biogas related issues
Production of heat or steam	Advantages of biogas cook stoves in wood scarcity and agricultural regions [106]; Life cycle energy and cost analysis of small-scale biogas facilities in rural regions [107]; Design features, construction material, feedstock, and operation parameters that made anaerobic digestion in small digesters [108]; Optimisation of techno-economic design of biogas digester [109].
Electricity generation or Combined Heat and Power (CHP)	Modelling and simulations of demand-orientated biogas-based power production [110]; Modelling of rural multi-energy complementary system with biogas cogeneration and electric vehicle considering carbon emission and satisfaction [111]; Load response of biogas CHP systems in a power grid [112]; Comparative analysis of different CHP systems using biogas for the cassava starch plants [113].
Vehicle fuel	Perspectives of biogas conversion into Bio-CNG for automobile fuel, Bangladesh case study [114]; Comparative analyses of the impact of biogas and biogas with hydrogen fuels on vehicle emissions and performance [115]; Sugarcane bagasse and straw as a biofuel to propel light vehicles [116]; System perspective on biogas use for transport and electricity production. Energies [117].
Fuel cell	Potential and constraints of solid Oxide Fuel Cells fuelled with biogas [33]; Biogas production and usage with legislations framework across the globe [15]; Biogas fed-fuel cell-based electricity generation: a life cycle assessment approach [118].

As per the IEA Report 2020 [32], about two-thirds of biogas produced globally goes for electricity and heat purposes, with an almost equal share of electricity and co-generation facilities. About 30% of biogas was consumed by the residential sector globally, while the residual share refers to biogas upgraded to biomethane, injected into a gas grid or used as fuel.

Small biogas systems are conductive in developing countries, where the rural population commonly depends on traditional biomass for cooking and heating. As the energy strategies commonly include small bioreactors, different governments and non-governmental institutions have supported their applications in Asia, South America and Africa. Nowadays, there are about 50 million small biogas systems [106,119,120]. Pilloni & Hamed [108] gave a review on design features, construction material, feedstock and operation parameters that make anaerobic digestion in small digesters attractive.

There is a special benefit in the possibility of the adjustment of conventional gas burners for biogas by changing the air-to-gas ratio. The only requirements of gas burners are a gas pressure of 8 to 25 mbar and a concentration of H_2S below 100 ppm to achieve a dew point of 150 °C [121].

The current increase in renewable electricity production requires power balancing, where demand-orientated combined heat and power production from biogas can be a suitable option. Grim et al. [110] analysed agricultural facilities, using cattle manure and sugar beet for biogas and CHP production. A developed dynamic production model showed that feeding management reduced the storage requirement due to the fast biogas production response to feeding. Furthermore, Ishikawa et al. [112] addressed the issue of load response with biogas CHP systems in power grid. They made an analysis of the systems operating in Germany and Japan that included load adjustment, start-up and shutdown behaviour, as well as exhaust gas emissions measurements during supply-demand adjustment. As concluded, biogas CHP systems have the potential to contribute to a long-term power equivalent adjustment, such as tertiary control reserve (TCR).

CHP systems use biogas and waste heat, resulting in much higher efficiency than using fuel to produce only power or heat. Gas turbines (in the range from micro to >100 kW) or internal combustion engines are commonly engaged. Such systems require removal of water vapour and H_2S (<100 ppm) from biogas [121]. Yin et al. [113] analysed energy demands for cassava starch plant. Results showed the ability to meet energy needs and economic feasibility for both the CPH system with reciprocating internal-combustion engine (ICE-CHP) and micro turbine (MT-CHP). Due to the lowest environmental impact, and the lowest and short payback period (4.57 year) ICE-CHP is suggested as first choice.

Although being intermittent, renewables are abundant in rural areas, and therefore a system that combines different energy sources can be recognized as a convenient option. Fan et al. [111] proposed a rural multi-energy optimisation model combining different technologies, where biogas cogeneration units improve system scheduling flexibility. The proposed model poses economic, environmental, and service benefits.

Future biomethane application relies on fuel cells as small-scale power plants of high efficiency exceeding 60% and low emissions. One of the largest digester/fuel cell units can be found at the South Treatment Plant in Renton, Washington, USA. About 4400 m^3/day of biogas produces 1 MW of electricity for site operation purposes [122]. Biogas can be converted into different renewable transport fuels, i.e., bio-CNG, liquefied biogas, syngas, and gasoline. A whole range of merits can be connected to the production of biogas as biofuels. However, the most emphasized are environmental benefits related to the reduction of GHG emission in the transport sector, generally from 60% to 80% compared to gasoline [106,114,123].

Shah et al. [114] gave an overview of available liquid and gaseous fuel commonly used as transportation fuel in Bangladesh. If the methane share in biogas is greater than 97%, biogas can be compressed at a pressure of 20–25 MPa to biomethane (Bio-CNG). Like regular CNG, bio-CNG shows high thermal efficiency due to a high octane number [124]. Special pertinence is that both petrol and diesel vehicle engines can be converted to bio-CNG through retrofitting with a gas tank and a gas supply system, in addition to the conventional petrol fuel system [125].

Biomethane can be liquefied (LBM). Achieved fuel has similar characteristics to LNG and is convenient for conventional vehicle engines. However, lower energy content of LBM (70% of gasoline) and required energy intensive methods, such as cryogenic technology, liquefaction, and pressure letdown, negatively impact the economics [114].

Syngas, as a mixture of hydrogen, carbon monoxide and a small amount of carbon dioxide, can be produced from pure biogas through reforming processes, such as dry reforming, steam reforming, and partial oxidative reforming, or any of their combination. Steam reforming is a widespread commercial process used to produce high purity hydrogen as clean transportation fuel [126,127].

Syngas can be converted into methanol and dimethyl ether and further upgraded into transport fuel like diesel, jet fuel or gasoline through the Fischer–Tropsch synthesis method [128]. Gasoline is produced by upgrading methanol through the methanol-to-gasoline (MTG) process. Biogasoline has a higher energy content and higher vapour pressure compared to biomethanol. Biogasoline can be directly used as transport fuel in a vehicle, or it can be mixed with biomethanol. Recently, partial oxidation has emerged as the most attractive and commonly used technology for biogasoline production

Karagöz [115] used a numeric engine model empirically validated by the global standard procedures for determining the levels of emissions and fuel consumption, LTC and NEDC. The research showed positive results when a low ratio of hydrogen (5% as molar) was added to biogas fuel. With the addition of H_2, the increase in fuel consumption dropped to 12.1% (compared to 16.1% in the case where pure biomethane is used). On the other hand, the increase in CO emissions decreased from 21.6% to 11.7% during the NEDC cycle.

Hydrogen derived from biogas can directly feed fuel cells. The reforming process can be succeeded by either internally employing fuel cells or externally with a catalytic pre-reformer. Abanades et al. [15] sees fuel cells as the cutting-edge application of biogas. Fuel cells utilization ranges from large-scale power plants to small scale portable power supply microelectronic equipment [129]. Although fuel cell-based power plants using biogas are not common in industrial application, more pilot projects are being initiated [33]. Their integration with other power generation systems like gas turbines or microgas turbines improve their performance. A biogas fuelled integrated solid oxide fuel cell (SOFC)-CHP represents a new efficient system that could surpass the issues of heat and power demands for decentralized grids [130,131].

4. Biogas Production in EUROPE

The EU has been recording an increase in the use of bioenergy, which is mainly due to the higher use of biogas rather than solid biomass. Biogas for electricity generation, heating and cooling took 8.3% (394.5 PJ) of the total bioenergy used in 2017, which means a five-time increase compared to 2005. Almost 60% of biogas is used for electricity production [132,133].

As for the size of the installed biogas plants capacity in 2017, the figure has tripled since 2005. Table 7 shows the total number of biogas and biomethane plants in the EU in the last decade. As seen, the number of biogas plants manifestly increased from less than 10,500 in 2010 to almost 19,000 in 2020 [134]. The increase in the number of facilities is accompanied by an increase in biogas production. As per the 19th EU Observ's Report [135], the European Union produced 16.8 Mtoe of biogas in 2018. In the meantime, since the UK withdrew from the EU, biogas production in the EU 27 was 13.8 Mtoe in 2018.

Table 7. Total number of biogas and biomethane facilities in the EU in the period 2010–2020 (according to [132,135]).

Year	**2010**	**2012**	**2014**	**2016**	**2018**	**2020**
Biogas facilities	10,508	13,812	16,834	17,432	18,943	Approx. 19,000
Biomethane	187	232	367	477	627	880

The Biogas barometer 2020 published the biogas production data by the EU member states in 2018. Analysis of published data is made and represented by Figures 5–7. Germany, UK, France and Italy are the leaders in biogas production in Europe (Figures 5 and 6). Analysis of the report data shows that Germany stands out as the most important producer, producing about 44% of the total biogas quantities in 2018. The United Kingdom participated with 17%, while Italy and France took shares of 12% and 4%, respectively. Among other countries, the Czech Republic and Denmark show a notable shift in production. Biogas from AD processes is most represented in the EU, participating with a high 84% of total biogas production. Germany, France and Italy produce a significant part of biogas by other AD processes, amounting to 98%, 79% and 62%, respectively, of their national biogas production (Figure 7). AD gas production in Germany far surpasses the production of other member states, amounting 52% of total biogas production by AD process in the EU. Landfill gas production is well represented in the UK, participating with 43% of total national biogas production. With regards to the EU, more significant landfill production can be found in Italy, France and Spain. Italy produces 27% of total landfill biogas in the EU, which is 18% of the country production. France participates with 24% of biogas from landfills, which is 34% of the country production. A high presence of landfill biogas locations can be found in Spain, where landfill production participates with 56%. Spain produces about 12% of landfill biogas in the EU. Biogas from sewage sludge is presented by less than 5% in total biogas quantities produced in the EU. Poland, Spain and the Netherlands take advantages of sewage sludge for biogas production. Sewage sludge participates with 40%, 25% and 18%, respectively, in their domestic production.

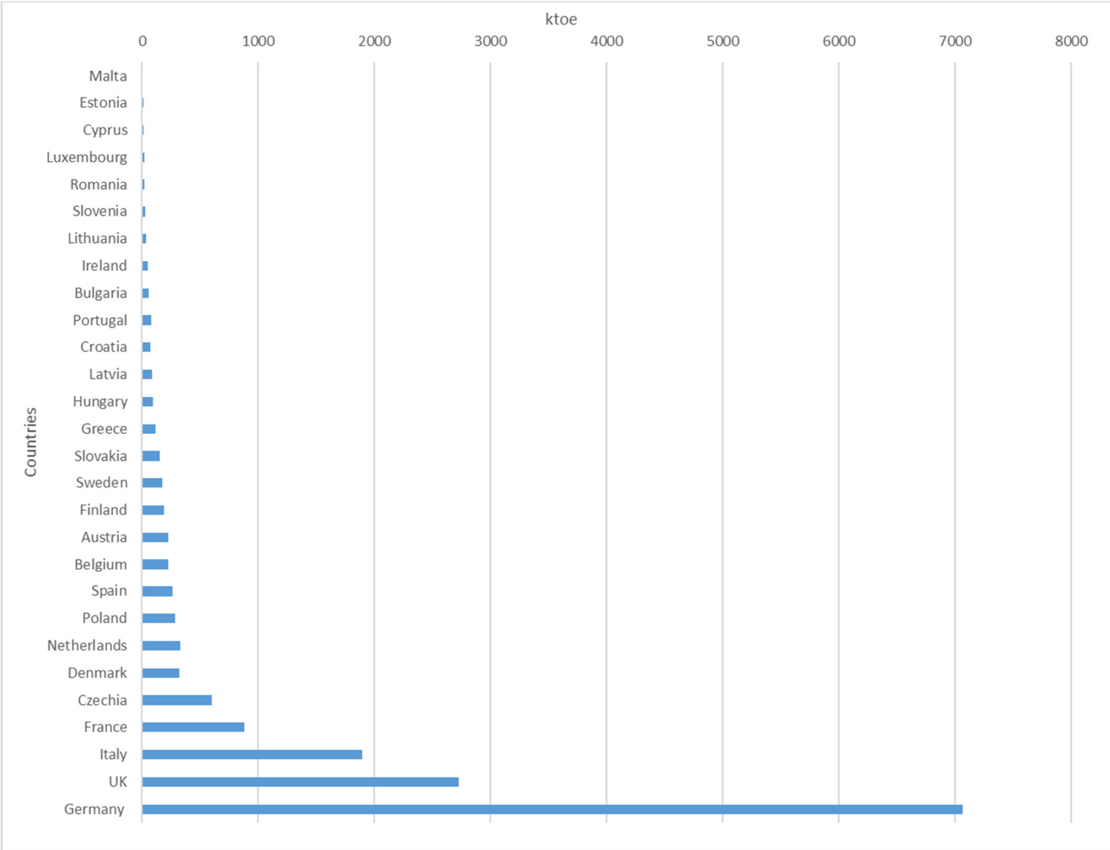

Figure 5. Biogas production in ktoe by countries (according to [135]).

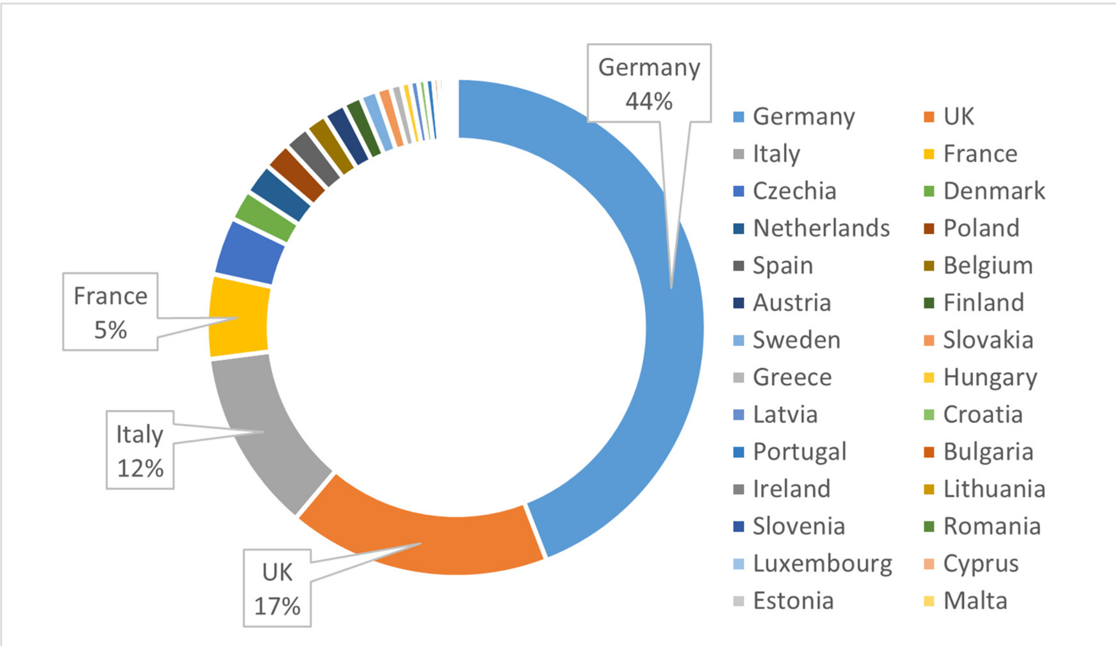

Figure 6. Related shares in biogas production (according to [135]).

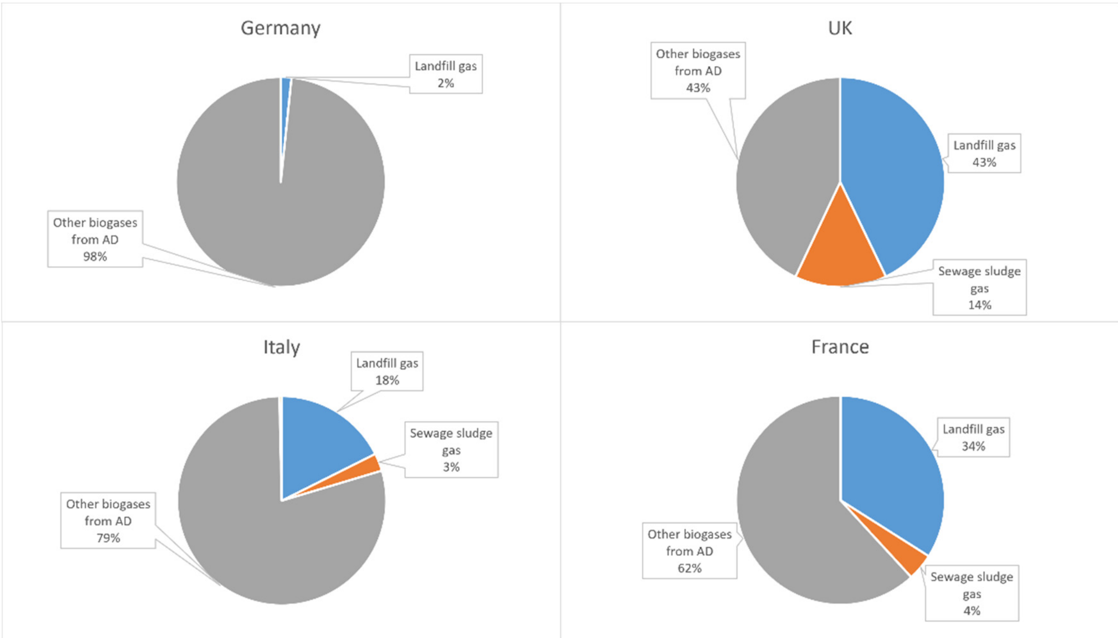

Figure 7. Leading countries in biogas production (according to [135]).

The increase in biomethane facilities is even steeper. In the last decade, the number of biomethane facilities in Europe have advanced from 483 plants in 2010 to 880 plants operating in 2020, reaching a total of 32 TWh of biomethane in Europe. Particularly significant progress can be seen in an only two-year period, from 2018 to 2020, when the number of biomethane facilities increased rapidly by more than 50%. There are currently 18 countries producing biomethane in Europe. Germany has the highest share of biomethane plants (232), followed by France (131) [136]

The Biomethane Map 2020 [136] published by the European Biogas Association (EBA) and the Gas Infrastructure Europe (GIE) shows data on infrastructure for biomethane production. Data analysis on substrate use was made and presented in Figure 8. It can be concluded that most of the plants installed in the EU use agricultural substrate as feedstock. Agricultural substrate (AGR), which includes agricultural residues, manure and plant residues represents the base for 28% of facilities. Energy crops (ENC) are used in 25%, while sewage sludge and waste (SWW), and bio and municipal waste (MWW) are feedstock for respectively 14% and 12% of the total biomethane facilities operating in the EU.

Production in Germany is highly dependent on energy crops. There, about 78% of crop-based plants are installed, while bio and municipal waste substrates have the second position (6% of installed facilities in Germany), slightly higher than other substrate alternatives. About 71% of biogas plants installed in France base their production on substrates coming from agricultural production except for energy crops, which includes manure, straw, cover/catch crops and crop residues. Sewage sludge facilities are represented by 14% in the EU, mostly applied in Sweden. Sweden has about 37% of the total number of such facilities installed in the European Union. Industrial organic waste from food and beverage industries (FAB) is the base for 5% of biomethane facilities, while landfilled waste based biomethane facilities (LAN) participate with only 2%. For about 2% of biomethane facilities there was no data (marked as UN in Figure 8) on the substrate presented on the Biomethane Map 2020 that was used.

Among other member states, growth in biomethane plants can be seen in Denmark and the Netherlands, having respectively 7% and 6% of the total number of installed plants in the EU. The Czech Republic has 38 facilities installed, which represent 5% of the total installed plants.

Substantial biomethane production is seen in non-EU member states, UK and Norway. In the UK, there were 80 biomethane facilities, mostly relying production on energy crops (60%).

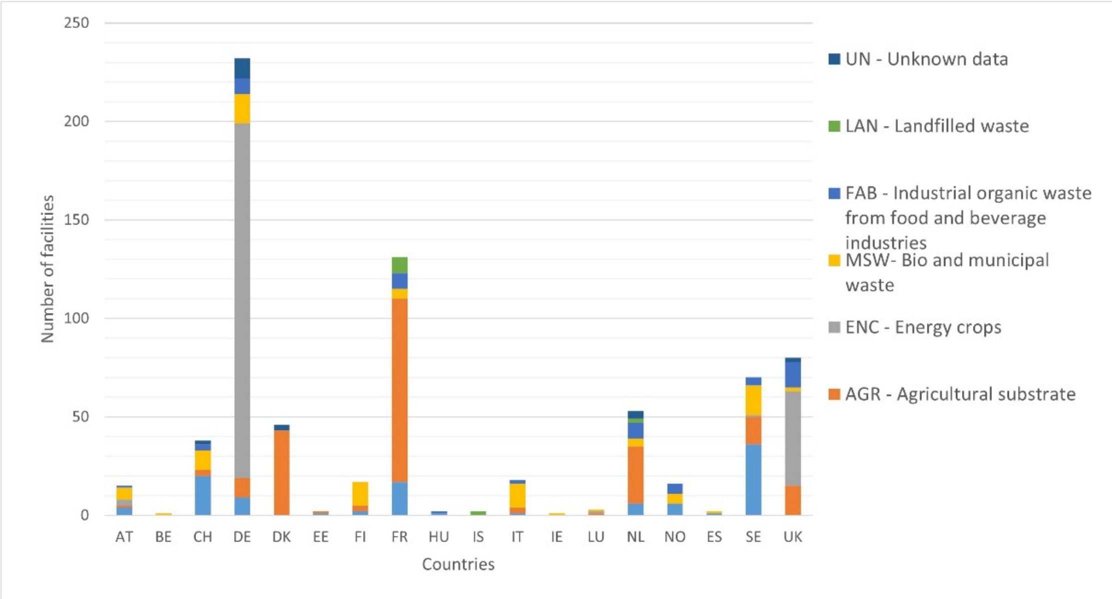

Figure 8. Distribution of facilities per feedstock in EU member states, UK and Norway (according to [136]).

The Biomethane Map 2020 [136] also shows data on infrastructure for biomethane production. Data analysis on the upgrading process used in 18 countries—biomethane producers in Europe, which means 16 EU member states, the UK and Norway were included and are presented in Figure 9. The facilities are grouped by upgrading the process used into chemical scrubbing (CHS), cryogenic separation (CRY), membrane separation (MEM), physical scrubbing (PHS), pressure swing adsorption (PSA) and water scrubbing (WAS). However, there is an evident lack of published data for some countries, which means that about one third of the data is missing (marked as UN data on diagram), mostly related to the UK and France. Since France and the UK belong to the European leaders in biomethane production, general conclusions cannot be made. Nevertheless, it can be seen that in other member states (data for France is not included) CHS and WAS processes are almost equally presented, with a light higher contribution of WAS (almost 20% compared to 19% of CHS). On the other hand, Germany, as the largest producer relies on CHS (34%), followed by WAS (27%) and PSA (21%). WAS is also highly present in Sweden with 61% of upgrading.

However, the same order in representation of individual technologies in biomethane purification processes in Europe is given by Niesner et al. [83], who stated that WAS was the most prevailing technology, applied at almost 40% of all biomethane plants.

MS appears as the main basis for biomethane production in the Netherlands, amounting to more than 69% or 42% in the Czech Republic. With regards to CRY technology, there are 2 facilities operating in the Netherlands and Sweden.

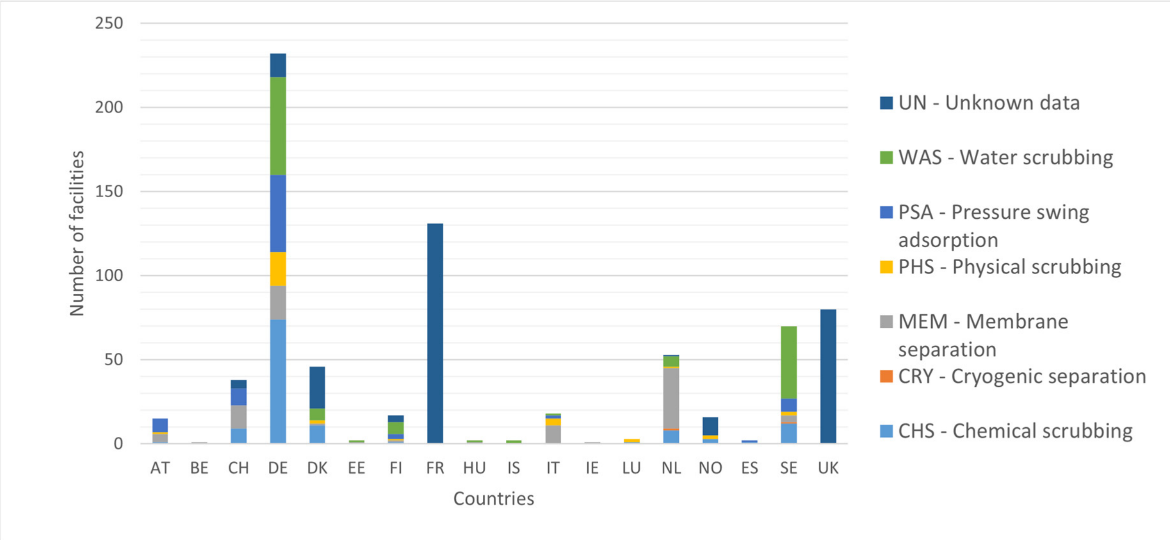

Figure 9. Distribution of facilities per upgrading process in EU member states, UK and Norway (according to [136]).

5. Discussion

In Europe, AD technology for biogas production was applied during the Second World War, when the lack of energy-initiated thoughts of switching to alternatives. Later, being less competitive than fossil fuels, biofuels were marginalized. A non-systematic approach to the biogas issue has resulted in its further neglect. Biogas solutions were just sporadically mentioned in the "Waste Directive" [137] and the "EU Bioeconomy Strategy" [138]. Recently, the situation has started to change. The EU targets adapted in the "REDII Directive" [4] affect biogas indirectly by requiring the share of energy from renewables of at least 32% and 14% of renewables in transport fuel to be achieved in 2030. Today, biogas production is a part of circular economy [139–141], with a central position in new European Green Deal strategies, such as the EU Methane Strategy [9], The Energy System Integration Strategy [10] and the "From Farm to Fork-Strategy" [11]. Furthermore, the recently proposed framework to decarbonize EU gas markets set the rules for the easier integration of biomethane in the gas grid with a legislative proposal to reduce methane emissions in the energy sector.

As per the EBA Statistical Report [15], membrane separation is still the leading upgrading technology, applied at more than 35% of biomethane facilities. The use of cryogenic separation is in its early development phase with 11 new plants in 2020. Currently, thermal gasification is experiencing an early commercialization phase, and hydrothermal gasification is being demonstrated with pilot projects located in France and the Netherlands [35,36,142]. Most conventional technologies applied to natural gas processing are efficient in biogas cleaning and upgrading. However, if separated CO_2 is vented into the atmosphere, the environmental benefit of the applied technology is questionable.

Due to that, biological CO_2 removal processes (e.g., microalgae-based CO_2 fixation, enzymatic CO_2 dissolution or fermentative CO_2 reduction) have to be the focus of future research [14,81,103].

Same "green trend" should be applied in each stage of biogas production. Process pretreatments, such as mechanical, microwave, thermal, chemical or biological, are available for both extending feedstock options and improving AD process efficiency. Today, due to the use of chemicals and waste liquid generation, some commonly applied pretreatment methods (hydrothermal, acid, alkali pretreatments) are not considered as green options. The feedstock for biogas production may contain crystallized cellulose and hemicellulose polymer matrix that is encrusted by the highly polymerized phenolic lignin causing difficulties in the conversion processes. Therefore, there is a need for other innovative pretreatment methods in order to facilitate biological digestion. Ecologically acceptable options are needed. Ultimate research showed that great biogas yield can be achieved if ionic liquid is used in the pretreatment process [74]. While high cost and immature recovery technologies limit its usage, electrochemical pretreatment of biomass appears as a promising tool for the utilization of lignocellulose.

While biogas production from locally available feedstock represents a generally sustainable energy model, special attention must be paid to the occupation of a huge surface of arable land with the large-scale production of crops intended for energy purposes. Therefore, new regulations are focused on incentives that favour the use of organic and farming waste. The first generation of biofuels with a high risk of indirect land use change (ILUC) when land purpose is converted from grasslands and forests to crop production, is going to be phased out by 2030.

Generally, biogas production and utilization are present in Europe and North America. There were about 1000 biomethane facilities operating worldwide in 2021. Besides the markets developed in Europe and the United States, the biomethane market is under development in Brazil, Canada, China and India [142].

Combined biogas and biomethane production in Europe exceeded 191 TWh of energy in 2020, and it is expected to be doubled by 2030, while by 2050, production can achieve over 1000 TWh). The great availability of biodegradable substrate ensures biomethane potential in Europe at the level of 1350 TWh [36]. Over the past decade, there was an increase of 90% in biogas industry (120 GW installed in 2019), and Europe contributes with over 70% of the world biogas generation. Developed countries have installed advanced large-scale facilities for utilizing biogas [15]. About two-thirds of biogas produced globally goes for electricity and heat purposes, approximately 30% is consumed by the residential sector, while a small residual share refers to biogas upgraded to biomethane, injected into the gas grid or used as fuel [31]. Biogas has usually been used in the production location vicinity for electricity and heat generation purposes. Produced electricity is usually consumed locally, or it can be handed over to the grid. Even the most promising forecast of biomethane upgrading considers consuming 60% of biogas in this way by 2030 [13], because high capital and operating costs were the main obstacles for more significant developments, especially when compared to the relatively low natural gas price. The natural gas price had stayed relatively low and stable for a decade. The current situation could be changed by the latest Ukraine crisis that dramatically affected the energy market. Although the need for renewables is well recognized in the EU energy strategy and action plans, strong dependence on the gas import from Russia requires faster actions in supply source diversification. This will certainly result in higher gas prices. In such circumstances, biomethane production can become a cost-effective option.

Process input varies significantly by region, since it is imitated to locally available materials that have acceptable transportation costs. Germany, UK, Italy and France are the leaders in biogas production in Europe. Biogas from AD processes is most represented in the European Union, participating with a high 84% of total biogas production, and agricultural-based biogas and biomethane facilities dominate production. Germany is deserving for the majority (52%) of AD biogas in the EU. Landfill gas production is well represented in the UK (43% of total national biogas production). More significant landfill production can also be found in the EU member states Italy, France and Spain. Biogas from sewage sludge is poorly presented by less than 5% in total biogas quantities produced in the EU. Sewage sludge practice is applied in Poland Spain and the Netherlands.

Biomethane facilities reached a production of 32 TWh in 2020 in Europe. There are currently 18 countries producing biomethane. Germany has the highest share of such plants (232), followed by France (131). Most of the European plants use agricultural substrate (28%), while the second position refers to energy crop feedstock (25%). Germany production relies on energy crops (78%) and biomethane production in France is highly dependent on substrates coming from manure, straw, cover/catch crops and crop residues (71%). Sewage sludge facilities participate with 14% in the EU, mostly applied in Sweden

[15,136]. While Germany, Great Britain, France and Italy stand out as the largest producers of biogas in Europe, most of the countries are in the early development stage. One of them is the Republic of Croatia, where biogas production and use is in its early phase, while biomethane production realization is planned for the future, as per the new national strategy for development of energy sector for the period 2030, with a view to 2050.

The first biogas plant in Croatia with installed power of 1 MW started to operate in 2009. According to the Report 2020 issued by Croatian Energy Market Operator (HROTE), in Croatia there are 41 biogas power plant with overall installed power capacity of 45.9 MW. Power plants with installed capacity of 1 MW are predominant. Commonly used feedstocks are manure (5–60%) and corn or grass silage (25–35%). The facilities are mostly located in the continental part of Croatia.

The analysis confirmed that biogas production and utilization differ significantly by region. Non-aggressive progress in the development of the biogas industry has been driven for a long time by specific states that recognized the value of biogas, primarily in Europe and North America.

Nevertheless, the implementation of adequate incentive is essential for the further development of the biomethane market. Irrespective of significant initiatives and the evident development of common biogas policy, it is obvious that specific instruments for biogas development stimulation may not be directly transferred to all countries since they are strongly dependent on regional and local conditions, mainly driven by energy system differences, but also the available substrate options.

6. Conclusions

The main advantages of using biogas and biomethane are its contribution to baseload power generation and reducing energy dependence. Biogas is suitable as a substitute for fossil fuels in power generation (both small and large scale), transportation and household sectors. Although the production of biogas is constantly increasing in European countries, the energy is mostly used for heat and electricity. The profit from biogas would be better if the biogas was purified and sold as fuel. In this way, less greenhouse gases are released into the atmosphere. Emission reduction can be considered from different points of view: Fossil fuel replacement, avoidance of methane emissions from animal or plant manure, production of green manure, etc. Overall, the benefits lead to a negative carbon footprint. However, the multiple benefits of biogas are not usually recognised as a value, resulting in the price of the final product being much higher than that of fossil natural gas. For this to change, the technologies for upgrading biogas to biomethane must be continuously improved. Recent geopolitical relations have led to instability in the natural gas market, resulting in the highest natural gas prices ever. This could also be the trigger for faster and greater inclusion of biogas and biomethane in the future energy mix.

Decarbonized Europe considers renewable electricity and renewable and low-carbon gases. Strong policy and regulatory support for the promotion of biomethane production and clarification of market rules are needed to achieve these ambitious goals.

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