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BIOGAS THROUGH ANAEROBIC DIGESTION FROM WASTE STREAMS AS A RENEWABLE TRANSPORTATION FUEL - A BRIEF REVIEW OF TECHNOLOGY

Biogas through Anaerobic Digestion from Waste Streams as a Renewable Transportation Fuel - A Brief Review of Technology

Author: Erik Karlsson - RISE Research Institutes of Sweden
Reviewers: Mats Edström, Daniel Tamm - RISE Research Institutes of Sweden

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Cover Image: Refueling of a gas car with biomethane, a commercially available renewable transportation fuel with about 90% lower greenhouse gas emissions compared to traditional fossil fuels.
Credit: Daniel Tamm

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ABSTRACT

Upgraded and purified biogas from anaerobic digestion, also known as biomethane, is an environmentally friendly renewable transportation fuel that is already commercially available for road transports. Biomethane shows low emissions of greenhouse gases compared to other transportation fuels, especially if waste streams are used as feedstock, and the emissions can even be negative in the case of manure. In this review, a technical overview is given for the available technology and how to process different organic waste streams and convert them to transportation fuel. The dry solids content (i.e. the viscosity) and particle size are the most important parameters determining the type of technology to be used: a sugar rich water stream is very different from agricultural crop residues. Another important aspect is the amount of contamination and unwanted material in the feedstock, since it is desirable to produce a high value digestate that can be used to fertilize agricultural land. The carbon dioxide separated from the biogas during the upgrading to biomethane can also be considered as a valuable by-product, and here there is room for development. It is also desirable to improve the productivity of the process by increased methane yield and higher energy efficiency, and there are opportunities for better overall resource efficiency and economy if the production is integrated with other processes.

BACKGROUND

With the European Green Deal, EU should reduce the emissions of greenhouse gases to zero by 2050. From the end user perspective, transportation is the largest emitter of greenhouse gases and needs large efforts to be converted to climate neutral technologies.[1] Conversion to biofuels is one measure, where biogas from anaerobic digestion of waste streams is one of the biofuels that give the largest reduction in greenhouse gas emissions compared to conventional fossil fuels. Depending on feedstock and assumptions, the reduction of emissions ends up at around 90%.[2,3] The reduction is especially high if using manure as feedstock: the emissions can then even be negative as methane emissions to the atmosphere from untreated manure are avoided.

Refueling of a gas car with biomethane, a commercially available renewable transportation fuel with about 90% lower greenhouse gas emissions compared to traditional fossil fuels. Credit: Daniel Tamm

To be used as a vehicle fuel, the raw biogas needs to be upgraded to pure biomethane (by removing carbon dioxide and impurities) and compressed, making it equivalent to compressed natural gas, CNG, so it can be used in conventional gas vehicles. For distribution of the gas, using existing gas grid is always the best choice, but if not available road transportation is also possible.[4] Liquified biomethane (equivalent to liquified natural gas, LNG) is emerging as a fuel for heavy-duty transport, both on road and by ships, as not many other renewable fuels are available for these applications.[5,6] There are also ideas of converting the methane to other types of fuels, e.g. Fischer-Tropsch fuels, methanol and hydrogen: these fuels are less mature and have lower potential in the short-term[7] but can be very interesting for production of aviation fuels[8].

Biogas has been produced for many years and has grown during the later years, now around 25 times higher than 1990, but still only corresponds to around 4% of the natural gas consumed in Europe.[9] Unfortunately, the total biogas production in Europe has levelled out slightly in the last years, but the conversion to upgraded biomethane is growing strongly and increased by 13% under 2018 to 23 GWh (compared to 65 GWh of electricity produced).[10] Europe is the leading continent and contributed to just over half of the global production.[11] Germany is one of the leading countries with the largest production in both total number and per capita in Europe, but this is to a large extent achieved by the use of energy crops.[10] Denmark, Sweden and Germany are the leading countries when it comes to upgrading to biomethane.[10]

AIM AND SCOPE

The aim of this literature review is to give a state-of-the-art overview of biogas production by anaerobic digestion from waste and residues (according to The Revised Renewable Energy Directive, REDII, Annex IX A) to transportation fuel (compressed or liquified biomethane). Energy crops as feedstock and utilization of the gas for heat, power or industrial use, even though also common, are not considered.

STATE-OF-THE-ART REVIEW

Figure 1 shows the basic steps in a biogas plant producing biomethane. When talking about biomethane production, the process is often divided into two types: wet and dry digestion. Wet digestion is the traditional technology where the feedstock must be pumpable. Dry digestion uses stackable feedstock and therefore relies on other technologies for transportation than pumps. The difference is further discussed under reactor design, but both methods use the same basic steps. Anaerobic wastewater treatment can also be used to produce biomethane, and here there is a potential to replace the aerobic treatment mainly used today. In addition to industrial scale anaerobic digestion, smaller farm scale plants are also common and have the advantage of needing less transportation of feedstock and digestate.

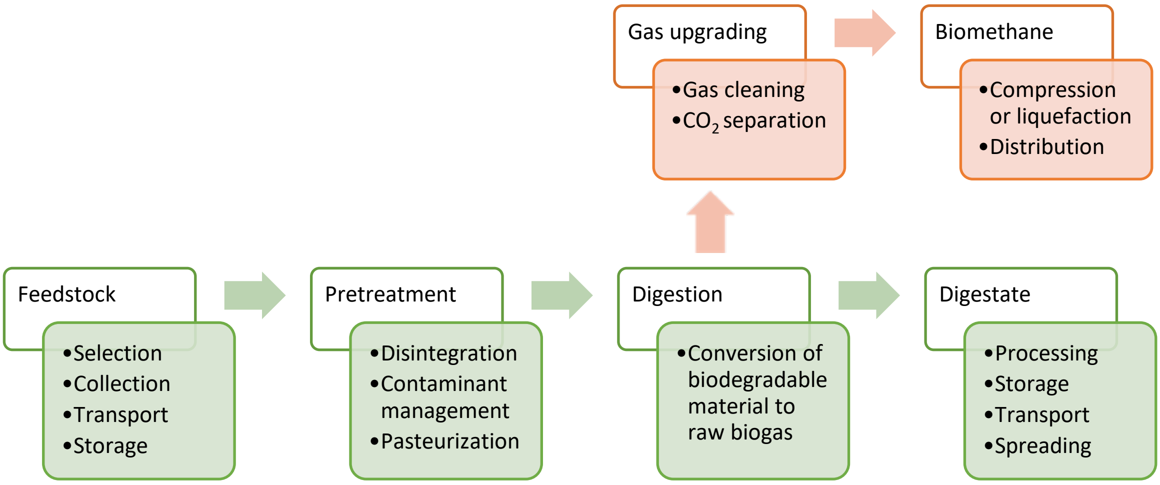

Figure 1. Overview of the different parts in a biogas plant, where each of them is explained individually in the subsequent sections.

WASTE STREAMS SUITABLE FOR ANAEROBIC DIGESTION

Feedstocks suitable for biogas production are easily degraded during anaerobic digestion, which means that they often have a high content of sugar, starch, proteins or fat, and a relatively high water content might be beneficial as well.[12] This is in many aspects the opposite requirement of thermochemical conversion (where dry lignocellulosic biomass with low ash content is preferred), and therefore the two technologies are complementary when it comes to utilizing all different biomass streams that are available.[12,13] The chemical composition of the feedstocks is normally complex as they are of biological origin, and the composition of waste streams are often varying, which makes an exact characterization almost impossible. Basic characterization is measurement of **dry solids content** (DS), **volatile solids content** (VS) and plant nutrients, and they are inexpensive to perform. For wastewater and diluted streams, it is more common to use **chemical oxygen demand** (COD), **biological oxygen demand** (BOD) or **total organic carbon** (TOC). **Biochemical methane potential** (BMP) test can be used to determine the potential methane yield of a feedstock, but requires batch digestion tests in lab scale, and is therefore more costly and time consuming. An alternative way that is faster is to search for BMP literature data for the feedstock. The feedstock might contain pollutants that needs to be considered as they affects the process and/or digestate quality, which are mainly of physical (e.g. plastics, metal and glass), chemical (e.g. heavy metals and persistent organic pollutants) and biological character (pathogens).[12]

Table 1. Summary of the main sectors providing waste and residual streams suitable for biomethane production. The most important examples of feedstocks for each sector are stated, together with the general advantages and disadvantages.

Feedstock source	Examples	Pros	Cons
Municipalities			
Wastewater treatment plants	Primary and secondary sludge	• Well known • Mostly utilized today	• Low methane potential • Can contain chemical contaminants and pathogens
Households, retailers and restaurants	Organic solid waste	• High availability • High methane potential	• Complicated logistics • Contains physical contaminants (e.g. plastics and metals)
Agriculture			
Livestock production	Liquid and solid manure	• Large quantities available • Cattle manure stabilizes the process	• Liquid manure has low methane potential • Solid manure can be difficult to process • Can contain pathogens
Crop production	Crop residues and surplus grass	• Large quantities available • High methane potential	• Needs collection and storage • Lignocellulosic rich material less digestible
Industry			
Food production	Wastewater, sludge and by-products	• Low degree of chemical and physical contamination • High methane potential from by-products	• Stringent legislation for animal by-products
Biorefineries (e.g. pulp, paper and biofuels production)	Wastewater, sludge and residues	• Large future potential • Integration opportunities with the biorefinery (especially heat)	• Lignocellulosic material less digestible • Toxic and inhibiting components might be present

The sectors with available waste and residual streams that can be used as feedstock, or substrate as it is often called, are mainly municipalities, agriculture and industry, where some examples are given in Table 1. Primary and secondary (also called bio) sludge from municipal wastewater treatment plants has been anaerobically digested for a long time but it has still a potential to increase the biomethane productions as the biogas is often not upgraded. Municipalities also generate food related waste streams from the whole chain of production, retailing and consumption, that have high methane potential and are only partly utilized for anaerobic digestion today.

There is a considerable potential for future increased biogas production from the agriculture feedstocks, where manure from livestock production may have the largest potential for biogas production. Residual streams from crop production as straw and tops but also catch crops and surplus grass can also contribute significantly.[14–16]

Industries processing food or biomass are also producing waste streams that are very suitable for biogas production: they are often homogeneous, easily digestible and rich in lipids, proteins or sugars.[12] In the strive for converting our society to a biobased economy, biorefineries will have a large potential for biogas production as many suitable residual streams will be generated.[17] There are also large opportunities to make the anaerobic digester and gas upgrading an integrated part of the rest of the biorefinery, where the bioethanol production is one of the most simple examples.[18,19] Furthermore, biogas plants can also deliver lignocellulose rich feedstock to biorefineries, extracted from the digestate, as the degree of conversion of the organic material in a biogas process is quite low for feedstocks rich in lignocellulose. The pulp and paper industry, especially Kraft pulp mills, has large sludge streams that, unlike most other wastewater treatment, are not anaerobically treated.[20] This is because they are difficult to degrade, but there is ongoing research to overcome the hinders as well as a full scale demonstration project called Effisludge.[20–22]

Finally, shifting from aerobic to anaerobic treatment of wastewater streams means a large potential for increasing biogas production (in addition to the substantial amount of electricity that is saved when turning off the aeration in aerobic treatment), even though the aerobic process is less sensitive and easier to control. Many industrial wastewaters are suitable for biogas production commercial installations are already available.[23,24] This is especially true for wastewaters from food industry due to high energy content and they are often easily digested, but this can depend from case to case. The pulp and paper industry has large wastewater streams that are treated aerobically today, especially in Kraft pulp mills, but successful installations exist in sulphite and recycling paper mills as their streams are easier to degrade.[20–22] Also for municipal waste waters, anaerobic treatment is possible and already used in tropical climate,[25,26] but might also be used in colder climate.[27]

It is important to know that the methane production often increases if different feedstocks are mixed (co-digested) in such a way that the microbiological activity is boosted.[28] For example, liquid manure from cattle has a stabilizing effect in the process and if combined with more energy rich feedstocks, the process will run smoothly at high production rate. It is also important to consider logistical challenges as planning and cost of transportation of the feedstock, and at the same time making sure they are suitable for the process. Digestate management also needs planning and transportation, as well as enough arable land to spread it on.[12,29]

PRE-TREATMENT AND PASTEURIZATION

One or several pre-treatment steps are often needed before the feedstock is fed into the anaerobic reactor. The pre-treatment has several purposes:[28]

1. Disintegration to decrease particle size and increase bioavailability
2. Inactivation of pathogens to stop spreading of different diseases
3. Remove contaminants that cannot be degraded or disturb the process
4. Change concentration to fit the process, i.e. thickening or dilution

The pre-treatment methods can be divided in physical, chemical, biological and combined.[30] Disintegration is normally performed by some kind of mechanical process, that increases the surface of the feedstock and thereby makes it more available for the biological process, which, in turn, increases the degradation rate and/or methane production. It will also stabilize the process and might be necessary for operational reasons (e.g. pumpability and reactor mixing).[12,29]

Heat treatment is imposed from legal requirements to kill potential pathogens, where EU regulation No 1069/2009 and 142/2011 demands a maximum particle size of 12 mm and pasteurization for at least 1 h at 70°C (or equivalent measures) for animal biproducts and food waste. A common solution for pumpable feedstock is therefore to have one or several pasteurization tanks before the digester, but pasteurization can also be performed in the digester (if a minimum guaranteed retention time is fulfilled that gives an equivalent treatment as 1 h at 70°C at the digester temperature) or after.[29] One might expect that this thermal treatment also improves the methane yield, but the data is not conclusional.[31–33]

For organic household waste and food waste containing contaminants such as packaging material, pre-treatment is needed to remove material that would otherwise disturb the process or decrease the value of the digestate as a fertilizer.[34] There are several suppliers for this kind of pre-treatment equipment, which normally consists of some kind of milling or shredding in combination with sieving and air separation. Hard material as sand, stone, glass and metal can damage equipment and needs to be separated as well, for example by gravity or magnets.[29]

Dilution, often with water, is common at wet anaerobic digestion plants with a high share of high DS substrates. Dewatering can be beneficial for diluted feedstocks as it will improve the process efficiency by increasing the retention time in the digester. There is a risk though that nutrient and organic material will be lost in the removed water.[28]

A great abundance of different pre-treatment methods that aim to improve the methane yield has been studied, but it is difficult to draw conclusions regarding how beneficial they are when considering the performance of the whole plant, as the pre-treatment should also have a low energy consumption, be wear resistant and cost efficient.[12,34] Especially methods for improving the degradation of lignocellulose have large potential as it is such a common material, but to be techno-economic viable, integrations with other biorefineries are often needed.[35] There have also been substantial efforts to improve degradation of sludge from waste water treatment plants by different pre-treatment methods with varying results.[34]

ANAEROBIC DIGESTION AND REACTOR DESIGN

Anaerobic digestion is a very complex biological process where several different groups of microorganisms are involved, and the different growth requirements have to be met for all these microorganisms to have a stable process.[36] The anaerobic degradation process can be divided into four basic steps comprising the different groups of microorganisms:[28]

1. Hydrolysis of complex organic material to smaller molecules (mono- and oligomers).
2. Fermentation of the smaller molecules to intermediary products (as alcohols and fatty acids).
3. Anaerobic oxidation of the intermediary products, requiring close collaboration with the microorganisms performing the last step.
4. Methanogenesis of hydrogen gas and carbon dioxide, or from some larger molecules such as acetate, to form methane and carbon dioxide.

The microorganisms need nutrients for their growth and function, both macronutrients such as nitrogen, phosphorous and kalium, and micronutrients such as nickel, cobalt, molybdenum, iron, selenium, and tungsten for methanogens, and zinc, copper, and manganese for the hydrolytic bacteria.[37] However, it is also important to not have too high concentration of compounds that can inhibit the growth. Ammonia is an example of an inhibitor that can decrease the methane production, even though high ammonia levels are also desirable as it will make the digestate more valuable as a fertilizer. Other important factors are temperature, oxygen level, pH and salt concentration. The microbial communities are adaptive and can to a certain extent adjust to changes of feedstock or conditions, but the most stable process is normally also the most efficient one. Co-digestion is normally also best as it gives a more balanced substrate composition also including macronutrients and micronutrients which stabilizes the process and enhances the methane production.[28] The best way to start-up a new digester is to inoculate with digestate from another biogas plant, preferably one that operates under similar conditions as this can give faster start-up and better end results. The feeding has to be started at low load and increased gradually while monitoring of digestion parameters to avoid overloading. It can take several months before target capacity is reached and the microorganisms have adapted to the new environment.[28]

Picture from the inside of a plug flow reactor. During operation, the agitator is rotating along a center shaft. Credit: Daniel Tamm

When designing and operating an anaerobic digester, there are some basic parameters to consider as **organic loading rate** and **retention time**. Organic loading rate is a measure of the reactor feeding rate and is often defined as kg VS from feedstock per m^3 reactor volume and day: a low rate gives low gas production while too high rate can result in overload and process failure. The retention time is important as it defines how long time the microorganisms stay in the digester, and as they grow relatively slowly, the retention time must be so long that they do not get washed out from the reactor before they have time to multiply accordingly. Usually at least 12 days are needed, but normally a retention time around one month is anyhow needed to utilize the biogas potential in the feedstock.[28] In practice, the chosen retention time will be an economical trade-off between utilizing the potential of the feedstock and the plant (i.e. operating and investment cost), while also considering aspects as methane release from the digestate (which is regulated in some countries). If the liquid and solid phase in the digester are separated, the **solid retention time** can be longer than the **hydraulic retention time** or vice versa.

There are many different reactor designs for anaerobic digestion, suitable for different feedstocks and conditions. They can, however, be divided into three main types, shown in Table 2, where the solids concentration in the feedstock is the determining parameter. For diluted systems mainly consisting of dissolved organic material, for example industrial or municipal waters, the hydraulic retention time needs to be short (hours) to avoid too large equipment and investment costs, but still the solid retention time needs to be long to keep the microorganisms in the reactor. Many different high-rate anaerobic solutions for this exist, where up-flow anaerobic sludge blanket (UASB) and expanded granular sludge bed (EGSB) reactors are widely used. They utilize the ability of the microorganisms to form granules with excellent settling properties and thereby stay in the reactor[26,28]. Anaerobic baffled reactors (ABR) work like a multistage UASB which makes the process more efficient. The advantages with high rate processes as UASB digesters is that they are robust and can handle large variation in hydraulic retention time and organic loading rates. In addition, the sludge production is low and easily dewatered. Constraints are that methane is partially dissolved in the effluent and causes losses to the atmosphere. Although the digestion process converting most of influent COD into biogas, the outflow requiring appropriate post-treatment to meet the wastewater discharge permits.[38] Anaerobic membrane bioreactors (AnMBR) add membrane separation to the reactor and therefore allow for complete sludge recirculation and higher effluent quality, but fouling of the membranes might be a problem.[39] Wastewater from industry in general have a low content of macronutrients and micronutrients and addition of these may be necessary. They can also contain to high concentrations of some compounds, where for example wastewaters from Kraft pulp mills are rich of sulphate that can result in a low specific methane production due to competing sulphate reducing bacteria producing hydrogen sulphide.[20]

Continuous stirred-tank reactors (CSTR) have been used for a long time to digest different waste streams and produce biogas which can be upgraded to biomethane. Here two CSTRs are used to digest organic household waste and different industrial waste streams.
Credit: Daniel Tamm

Wet digestion is the most common technology and has been used for many years for example for the treatment of municipal sewage sludge. The condition for it to be used is that the material inside the digester (and hence the digestate) is pumpable and allows mixing (DS around 10% or less).[40] Due to the degradation in the digester, pumpable feedstock mixtures with slightly higher DS can be used (DS < 15%), but if solid feedstocks are used they normally need to be diluted.[29] A wet digestion plant normally has a simple design consisting of a continuously stirred tank reactor (CSTR) that, if correct pre-treatment of the feedstock is used, gives few (mechanical) issues. The most common problems are related to mixing, where accumulation of sediments in the reactor decrease the active volume, and floating layers detain material from contact with the microorganisms and complicate level control. Pumps are also sensitive to abrasive material. Otherwise the process is often stable and economical.[29]

Table 2. The three main types of reactor designs for anaerobic digestion, determined by the concentration of solids in the feedstock.

	Diluted systems	Wet digestion	Dry digestion
Feedstock type	Waste and process waters	Pumpable slurries	Stackable feedstocks
Approximate DS*	<2%	2%–15%	>15%
Reactor type	UASB, EGSB, ABR, AnMBR etc.	CSTR	Batch or plug flow
Temperature	Psychrophilic–mesophilic	Mesophilic–thermophilic	Mesophilic–thermophilic
Hydraulic retention time	Low	Intermediate–high	Intermediate–high
Solid retention time	Intermediate–high	Intermediate–high	Intermediate–high
Phases	Two	One	One or two
Characteristics	Robust and can handle high feed rates	Versatile technology e.g. suitable for co-digestion of liquid and solid feedstocks.	Newer technology that has advantages in some applications.

**The numbers are taken from SGC[41] and should be seen more as indications as the structure and viscosity can be very different between different feedstocks despite the same DS. For diluted systems it is more common to use COD or TOC instead of DS.*

Dry digestion, or solid-state digestion as it is also called, is suitable for solid or stackable feedstock (e.g. food waste and solid manure). The advantage is the high concentration in the system, reducing the reactor volume and transportation of digestate.[42] The system is also less sensitive to contaminants as plastic, metal, sand etc. as the material handling is adapted to a high content of solid material.[43] Dry batch reactors have the advantage of being very simple and robust, making it suitable for small scale or mechanically difficult feedstocks, but manual handling of feedstock and digestate makes the operational costs higher compared to other technologies.[29] The reactors are often similar to garages with gas tight doors, that are loaded and unloaded using a front loader. Several batch reactors are normally operated sequentially to maintain a constant gas production.[12] A liquid phase, leachate or percolate, is drained in the bottom of the reactor and showered on the top of the feedstock for heating and to improve mixing. Continuous plug flow reactors have the advantage of having a continuous gas production, little manual handling and a potentially high gas yield, and there are different designs with and without mixing and with horizontal or vertical flow.[44] The dry digestion technologies are being implemented in full-scale more and more from several suppliers[45,46] due to the advantages when digesting solid feedstocks.[47,48] Still, more work is needed to improve the efficiency, where the contact between the feedstock and microorganisms is an important challenge.[49] Due to the higher concentration, the risk of inhibition of the microorganisms increases during dry digestion, for example by ammonia.[47]

Dry digestion technology in plug flow reactors is the best available technology for some types of feedstock. Here two plug flow reactors are used at a biogas plant that produces biomethane for vehicle fuel out of organic household waste.
Credit: Daniel Tamm

Reactor temperature is a very important parameter to consider when designing the process and there are three different temperature intervals that are used:[36]

- Psychrophilic (<25°C)
- Mesophilic (37-40°C)
- Thermophilic (50-55°C)

Mesophilic and thermophilic conditions are the most common during anaerobic digestion, but intermediate temperatures as 41-45°C are also used sometimes.[36] When choosing the temperature, both the methane production and the heating demand of the process need to be considered. Mesophilic conditions limit the heating demand but still have a stable process with high gas production. Thermophilic conditions can, however, have even higher gas production but are often considered less stable and more sensitive for inhibition, especially by ammonia as the equilibrium with the nontoxic ammonium is shifted towards ammonia at higher temperature.[28] Thermophilic reactors with appropriate minimum retention time have the advantage of efficiently killing pathogens, which simplifies the system and might then have a lower heating demand than a mesophilic plant with pasteurization pre-treatment.[47] Psychrophilic systems have the advantage of needing no or very little heating, but the methane production rate is, on the other hand, significantly lower: it has been reported that about the double retention time is needed to give the same gas production as during mesophilic conditions.[50] Psychrophilic conditions are therefore mostly suitable for diluted systems (as they can allow a low hydraulic retention time and it is costly to heat a diluted stream)[51] and low-cost systems as covered lagoons[29], but potentially also for feedstocks with high ammonia levels[52].

The most common solution is to have the whole degradation in just one reactor, but it might be beneficial to divide the process into two steps: one reactor for acidogenesis (step 1-2 above) and one for methanogenesis (step 3-4 above). Then each reactor can be optimized individually to fit the needs of the respective microbial groups, which often results in fast and efficient biogas production in the second stage, but the division will never be complete, so biogas will be produced in the first reactor as well.[28] For wet systems, a retention time of 1-3 days is often recommended in the first reactor, and thermophilic conditions might be beneficial depending on feedstock.[53] In some contexts, these small reactors are considered as a pre-treatment which might be confusing. The dry batch process can benefit considerably from a two-step configuration as it is already separated into two phases: it then consists of a leaching bed which can be optimized for hydrolysis, while the leachate is digested separately in a high rate digester for diluted systems as UASB to improve methane production.[54]

Finally, it should be mentioned that process monitoring and control are crucial for having a successful operation. First off all, the temperature in the reactor should be kept as constant as possible and preferably not change by more than ±0.5°C.[28] The biological process is very complex and there is always a risk of imbalance between the different microorganisms. Especially the microorganisms in the last step, methanogenesis, are sensitive: if they stop thriving, intermediate products formed earlier in the chain start to accumulate, for example volatile fatty acids (VFA), leading to a decreasing pH which in turn makes the living conditions for methanogens even worse. Therefore, it is important to monitor parameters such as pH, alkalinity, VFA content, ammonia content, gas production and methane content, and take measures before the disturbances becomes irreversible.[28] Unfortunately, many of these parameters are difficult or expensive to measure online. There is progress in the field, but still development of inexpensive and reliable online measurements of digester parameters and feedstock characteristics would improve supervision and could facilitate the use of advanced AI based control systems.[55]

DIGESTATE MANAGEMENT

Large volumes of digestate are produced during anaerobic digestion, and a good management and utilization is important for both environmental and economic reasons. The nutrients and carbon in the digestate are valuable for farmers, especially if organic, but there are several factors to consider when spreading it on farmland:[12,28,56]

- Digestate quality (e.g. content of heavy metals, antibiotics, organic pollutions, pathogens and plastics)
- How much can be spread per hectare (determined by the amount of nitrogen and phosphorus)
- When to spread (when there are growing crops that can absorb the nutrients)
- Logistics (distance to the farmland, roads, storage, spreading technique etc.)
- Odour

Many of these factors are also regulated to minimize emissions to water and air that can cause eutrophication and global warming or pollute groundwater.[56] Since spreading is mainly performed during summer, large storage capacity of digestate is needed, and during storage leakages and emissions should be avoided as well. It is important to mix the storage carefully before spreading the digestate as solid material will sediment.[28]

Due to the high content of water in digestate, the transportation with respect to the nutrient content is costly. At the same time, the amount that can be spread per area is limited, often resulting in long transports for spreading.[56] For large scale biogas plants, it might therefore be interesting to concentrate the digestate in some way. Processing of the digestate is also interesting if the digestate has quality issues (e.g. contains impurities such as plastics), but in practice the quality of the digestate is mainly controlled by only using high quality feedstocks.[12] Digestate processing normally starts with the separation of the solid and liquid phases. For digestate rich in fibres, screw presses are most suitable, otherwise a decanter centrifuge is common, and flocculation or precipitation agents can be added to improve the separation.[12] The solid fraction can then be further processed for example by drying or composting. The liquid fraction still contains nutrients and organic material, where the relatively high content of nitrogen can be interesting to recover e.g. by struvite precipitation.[57–59] Further concentrating by evaporation or membrane filtration is also possible.[60] However, digestate processing is mainly economically relevant for very large plants with limited land for spreading of unprocessed digestate.[61]

The storage of the digestate can be solved in different ways depending on situation, e.g. stored at the plant site or closer to the farm land.[56] The recommendation for whether the storage tanks should be covered or not to prevent methane, nitrous oxide and ammonia emissions depends on the situation and local regulations, where the on-site storage are often covered while satellite storages closer to the fields are more commonly opened, but a cover also has the advantage of preventing rain from diluting and taking up storage volume.[12,28,29,56] The biological activity should be minimized during storage, but sometimes part of the storage is constructed as a post-digester to recover more of the remaining methane potential and then the biological activity should continue.[28] The gas production from storage and post-digestion varies, but figures between 1 and 10% of total gas production are common, where the temperature in the storage tanks is very important.[12,62] One approach to actively inactivate the biological activity and decrease emissions to air is acidification of the digestate to pH around 5.5, which also shifts the equilibrium between ammonia and ammonium towards ammonium that cannot evaporate. This will result in reduced emissions to the atmosphere of greenhouse as methane and nitrous oxide (N2O) but also ammonia (NH3).[63]

GAS UPGRADING AND DISTRIBUTION

A biogas plant is normally equipped with a gas storage to even out production and consumption, and when upgrading the gas it is enough to store a 2-3 hours of production or less.[12] For CSTR, the common solution is to store the gas inside the digester, and here the roof consists of a double membrane that allows changes in the volume. For other types of reactors, external storages made of flexible structures such as membrane cushions or detached double membrane globes are most cost efficient. [12]

The raw biogas mainly comprises of methane (50-70%), carbon dioxide and water, but also lower concentrations of impurities such as nitrogen gas, oxygen gas, hydrogen sulphide, ammonia, siloxanes, volatile organic carbons and particles.[12] These impurities can disturb the subsequent processing and utilization of the gas, for example by causing corrosion. Removing the carbon dioxide is called gas upgrading and, depending on which technology used, the impurities are removed before, during or after the upgrading process. The main technologies used for upgrading of biogas are presented in Table 3, where the market is dominated by water scrubber (largest share), chemical absorption, membrane separation (strong increase of share recent years) and pressure swing adsorption.[64,65] In addition, the different technologies can also be combined.[66] All technologies have pros and cons, and the site specific conditions affect which is the most suitable. For example, the required pressure after upgrading can be important for decision, where membranes are more suitable if the gas is intended to be distributed at high pressure. Cryogenic separation is a new technology, with just a few commercial installations, suitable for production of liquified biomethane. Upgrading to biomethane is normally not economical for smaller biogas plants due to the high investment cost.[64]

It is important to also remove the impurities present in the raw biogas, but exactly how this is done depends on the upgrading technology used and the concentrations that are present. To avoid malfunction of the gas vehicles, the dew point of the water in the biomethane needs to be significantly below the freezing point, meaning that condensation during compression and cooling of the gas is not enough: normally an adsorption process (e.g. temperature swing adsorption) is used for final drying.[67] In this step, even some particles, siloxanes, hydrogen sulphide and ammonia will be dissolved in and removed with the water, which has to be considered when designing systems for disposal of the water.[12] Adsorption on activated carbon can be used for lower concentrations of volatile organic carbons, siloxanes and hydrogen sulphide. Sometimes the raw biogas contains relatively high contents of hydrogen sulphide, and then biological oxidation can be used for removal, either in the digester or afterwards in the gas system. For some setups, there is a risk of contamination of the biogas with nitrogen and/or oxygen gas because air or oxygen needs to be added to the biogas. Hydrogen sulphide can also be removed in the digester by precipitation with iron salts.[64]

Off gases, i.e. gas streams from the gas upgrading containing the separated carbon dioxide, may need treatment before emitted to the air, depending on the upgrading technology. Here, it is mainly the residual methane that needs to be removed, for example by thermal or catalytic oxidation, in order to avoid unnecessary greenhouse gas emissions. Upgrading by chemical absorption, membranes (if several stages) and cryogenic separation produce relatively clean carbon dioxide streams, that can be interesting to purify and sell as an additional product.[66] There is also ongoing development work to produce hydrogen gas from renewable electricity and use it to convert the carbon dioxide to biomethane through microbiological or thermochemical methods.[66,68,69] This can also help balancing the electricity grid if the hydrogen gas production is allowed to vary depending on electricity price.

Table 3. The main technologies for upgrading of biogas to biomethane, where cryogenic separation is used for production of liquified biomethane whale the others are used for compressed biomethane.[64,66,69,70]

	Separation method	Pros	Cons
Pressure swing adsorption	CO_2 adsorption onto e.g. activated carbon, zeolites and carbon molecular sieves	• No chemicals needed • Separation of some N_2 and O_2	• Can have high methane loss • Batch process
Water scrubber	CO_2 absorption into water	• Simple technology • Low investment cost	• Potentially high water demand that also needs treatment • Potential problems with biological fouling
Physical absorption	CO_2 absorption into organic solvent e.g. polyglycol dimethyl ethers	• High absorption rate and efficiency • Separation of H_2S, NH_3 and H_2O	• Handling of organic solvents • Small amount of 50-80°C heat needed
Chemical absorption	CO_2 chemically bound to an amine in water solution e.g. ethanolamine (MEA) No compression of the gas needed	• High methane purity • Low methane loss • Low electricity demand • Separation of H_2S and NH_3	• High investment cost • High demand of 95-140°C heat • Foaming, amine loss and corrosion
Membrane	Separation through CO_2 permeable membrane with one or several stages at 10-20 bar	• Simple technology • Compact construction • No chemicals needed • Scalable	• Requires thorough cleaning of the gas to remove impurities • Membranes are sensitive
Cryogenic separation	Condensation or sublimation of CO_2 with very high pressure around 40 bar and substantial cooling <-45°C Used for liquified biogas	• High methane purity • Low methane loss • Low extra energy if production of liquid biomethane	• Highest investment cost • Highest electricity consumption • Less proven technology

There are two ways of distributing the biomethane from the biogas plant to the filling station: via gas networks or road transport. It is very beneficial to inject the biomethane into an existing natural gas network if available, but to allow this, the biomethane needs to fulfil certain quality requirements. The requirements differ between countries, and over and above purifying the methane, addition of petroleum gas (propane) might be needed to increase the heating value. The required pressure depends on which level in the grid the gas is injected to: a distribution grid with a low pressure of 30–100 mbar, a regional distribution network with a medium pressure of 4–16 bar or a supra-regional gas transmission network with high pressure of 32–120 bar.[12] In some situations far away from the natural gas grid, it can be economical to build a small local network for the biomethane distribution, but otherwise the gas has to be compressed or liquified and stored in mobile storage units that can be transported on road, which is normally more costly than using existing pipelines. The energy needed for liquefying biomethane is more than double the energy needed for upgrading, but liquefied biomethane is still more efficient for long distance road transports.[4] For security reasons, compressed biomethane needs to be odorized before distribution. To fuel the gas vehicles, the biomethane needs to be compressed to around 230 bar.[4]

PROCESS EFFICIENCY AND INTEGRATION OPPORTUNITIES

Utilization of waste streams is one way to improve resource efficiency and sustainability, and from those streams suitable for anaerobic digestion, biomethane can be produced as described in this review. In addition to improve the efficiency of each unit operation in the process, the overall resource efficiency and economy can be improved even further if material and energy streams are integrated with other adjacent processes to produce multiple products.[71] For example, anaerobic digestion can constitute the main platform of different waste based biorefinery concepts[72,73] or be a complementing part of other types of biorefineries[17,74,75]. In addition, biorefineries, as well as food industry, often have the advantage of having excess heat that can be used in the anaerobic digestion and thereby save energy.[76] The integration with agriculture is obvious when it comes to feedstocks and nutrient management, but integration in terms of energy is also possible (e.g. provide transportation fuel or heat integration with other parts of the farm).[77–79]

One of the most important aspects to improve the process efficiency of the biomethane production is of course to maximize the yield of the plant, which involves substrate selection and co-digestion, efficient pre-treatment and optimized reactor operation. These aspects have been discussed in previous parts and are constantly relevant research topics. For energy production, the balance between produced and internally consumed energy is important, and the energy efficiency of the process should be as high as possible. Compared to some other energy conversion processes, however, it is challenging to both define and quantify energy efficiency for anaerobic digestion, for example due to the fact that the energy content in the feedstock and how to include the agricultural system is not clearly defined.[80] The variations between different feedstocks and production conditions are also large.[81] The parasitic energy demands are mainly transportation (of feedstock, digestate and potentially biomethane), pre-treatment (disintegration, pasteurization etc.), reactor operation (heating, mixing and pumping) and gas upgrading (separation of carbon dioxide and impurities, and compression).[12] Unfortunately, mainly tractors and trucks driven by fossil diesel are used for transportation. The need of transportation is strongly dependent on feedstock and local conditions: anaerobic digestion of industrial wastewater might need no transportation at all, while organic household waste from the countryside might need long transportation. To minimize transports, location and logistics of the plant should be considered with great care. Some feedstocks need no pre-treatment, while organic household waste and some agricultural wastes need pasteurization and/or substantial processing to remove unwanted material (i.e. considerable demand of both heat and electricity). Heat recovery equipment, operating temperature, insulation and design of the reactor determines its heat demand, while pumping and mixing needs electricity. As described earlier, depending on the upgrading process, the ratio between heat and electricity demand varies, but compression will always have a high electricity demand.[12] Efficient equipment and design should be used in the plant to minimize electricity consumption.

As already mentioned, the best way to supply heat is by integration with other processes, because then the heat can be cascaded and reused more efficiently. The value of the heat is determined by the temperature, and here the anaerobic digestion has the advantage of only needing heat at relatively moderate temperatures (70°C if pasteurization, otherwise 35-55°C) at which excess heat is often available from other processes. If chemical absorption is used for biogas upgrading, there is however also need for temperature above 95°C. Another alternative for heat supply, depending on local availability, is district heating. Otherwise, the heat must be produced on-site. When biogas is used for combined heat and power production, there is normally more than enough heat to cover what is needed for the digestion process.[12] When producing biomethane, it is still possible to use a part of the biogas in a boiler or e.g. a micro turbine (which can also be used to treat any off-gases from the upgrading), but then there will be less of the valuable end product produced. If the plant is connected to the natural gas grid, it is common to use natural gas for heating, but this is less beneficial for the climate. A boiler for solid fuels, such as wood chips or pellets, can also be used. There are also suggestions to use solar heat for at least part of the heat demand.[82,83]

Heat integration or heat recovery within the biomethane production process, e.g. between the pre-treatment, digester and upgrading, is important to improve the energy efficiency.[84,85] Depending on the reactor configuration (Table 2), heat recovery is more or less difficult: In diluted systems, heat exchanging of feedstock and digestate is relatively easy, while in dry digestion it is almost impossible. On the other hand, solid material has a much higher energy density which increases the plant efficiency and makes heat recovery less important. In some situations, the diluted systems are not even heated due to the high parasitic energy demand and no excess heat available, but insulation of the reactor and piping is beneficial if the wastewater is already warmer than the surrounding.[86] In wet digestion plants with pasteurization of the feedstock, the hot feedstock (70°C) from pasteurization needs to be cooled before entering the reactor, which is done by heat exchanging with the incoming feedstock.[87] This is an important heat recovery operation, but heat can also be recovered from the digestate by for example pre-heating the feedstock. The challenge for the heat recovery is, however, low efficiency and malfunctions of the heat exchangers: sludges and slurries are viscous and contain particles giving low heat transfer coefficients as well as clogging and fouling. The heat exchangers used today are specially adapted to the situation, but the operation is still challenging. There are ideas of improving the heat transfer by scraping surfaces[88] or new geometries[89], but the investment cost must not be too high as the value of the recovered heat is relatively low. Struvite ($MgNH_4PO_4 \cdot 6H_2O$) is a mineral that often precipitates, causing malfunction of the heat exchangers and demanding regular acid cleaning. However, controlled precipitation can also be a way to create a valuable nutrient stream.[90,91]

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