

Biogas upgrading technologies – developments and innovations

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Task 37 - Energy from biogas and landfill gas

IEA Bioenergy aims to accelerate the use of environmental sound and cost-competitive Bioenergy on a sustainable basis, and thereby achieve a substantial contribution to future energy demands.

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Task 37 has previously published the following brochures about biogas upgrading:

- Biogas upgrading to vehicle fuel standards and grid injection, 2007
- Biogas upgrading and utilization, 2000

The publications are available on www.iea-biogas.net.

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Content

Introduction	4
Composition of biogas	5
Cleaning of biogas	6
Removal of water	6
Removal of hydrogen sulphide	7
Removal of oxygen and nitrogen	8
Removal of ammonia	9
Removal of siloxanes	9
Removal of water	9
Removal of particulates	9
Full scale technology for biogas upgrading	9
Pressure Swing Adsorption (PSA)	9
Absorption	10
Membranes	11
Comparison of different upgrading techniques	12
New developments in upgrading technology	12
Cryogenic upgrading	12
In situ methane enrichment	14
Ecological lung	14
Removal of methane from the off-gas	15
List of upgrading plants	16
List of biogas upgrading plant providers	18
References	19
Abbreviations and definitions	19

Introduction

Biogas is produced during anaerobic digestion of organic substrates, such as manure, sewage sludge, the organic fractions of household and industry waste, and energy crops. It is produced in large scale digesters found preliminary in industrial countries, as well as in small scale digesters found worldwide. Biogas is also produced during anaerobic degradation in landfills and is then referred to as landfill gas. The worldwide biogas production is unknown, but the production of biogas in the European Union was estimated to be around 69 TWh in 2007. The biogas production in the European Union has steadily increased over the last years (Fig. 1).

Biogas consists mainly of methane and carbon dioxide and it can be utilized as a renewable energy source in combined heat and power plants, as a vehicle fuel, or as a substitute for natural gas. The methane in the biogas can also be utilized in industrial processes and as a raw material in the industry.

Production and utilization of biogas has several environmental advantages such as:

- It is a renewable energy source.
- It reduces the release of methane to the atmosphere compared to e.g. traditional manure management or landfills.
- It can be used as a substitute for fossil fuels.
- A high quality digestate that can be used as a fertilizer is produced simultaneously with biogas.

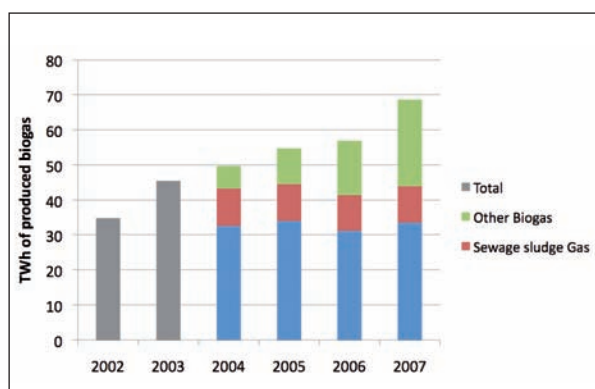


Fig. 1. Biogas production in the European Union between 2002 and 2007 (Biogas Barometer 2004–2008).

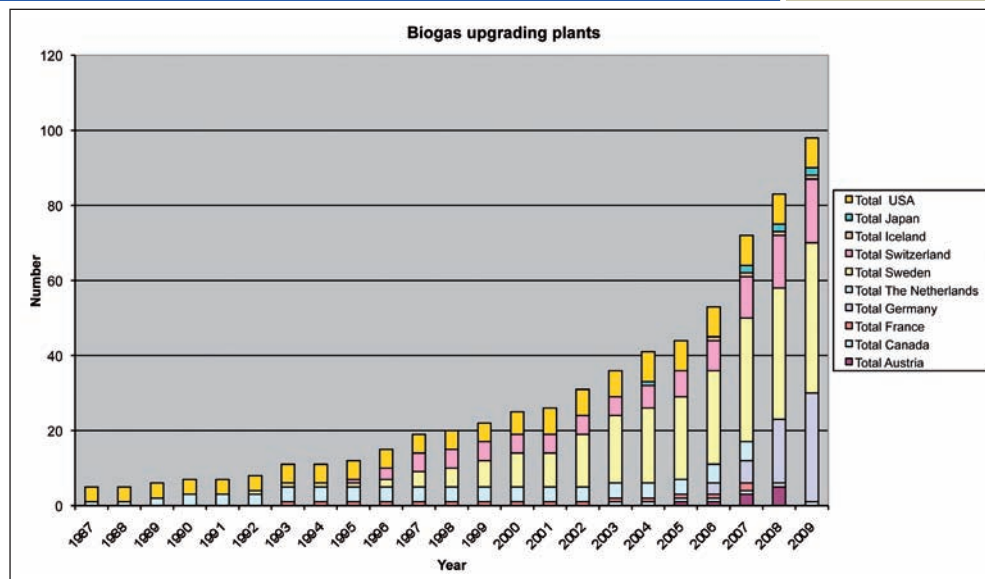


Fig. 2. Total number of upgrading plants from 1987 to 2009.

Depending on the end use, different biogas treatment steps are necessary. For some applications, where it is important to have a high energy content in the gas, e.g. as vehicle fuel or for grid injection, the gas needs to be upgraded. The energy content of biogas is in direct proportion to the methane concentration and by removing carbon dioxide in the upgrading process the energy content of the gas is increased.

Upgrading of biogas has gained increased attention due to rising oil and natural gas prices and increasing targets for renewable fuel quotes in many countries. New plants are continually being built. The number of upgrading plants was around 100 in 2009 (Fig. 2).

The process of upgrading biogas generates new possibilities for its use since it can then replace natural gas, which is used extensively in many countries. However, upgrading adds to the costs of biogas production. It is therefore important to have an optimized upgrading process in terms of low energy consumption and high efficiency giving high methane content in the upgraded gas. It is also very important to minimize, or if possible avoid, emissions of methane from the upgrading process, since methane has a greenhouse gas effect 23 times greater than that of carbon dioxide. This means that the methane content in the reject gas, in the water from a water scrubber, or in any other stream leaving the upgrading plant should be minimized.

Several techniques for biogas upgrading exist today and they are continually being improved. In parallel, new techniques are under development. These new developments, both for new and more traditional techniques, can lower investment costs and operational costs. The developments can also lead to other advan-

tages such as lower methane emission which is important from both an economical and environmental perspective. In this brochure the latest developments in biogas upgrading are reviewed.

This publication is produced by IEA Bioenergy's Task 37: "Energy from biogas and landfill gas". More information about the group as well as publications can be found on www.iea-biogas.net.

Composition of biogas

During anaerobic digestion (i.e. digestion in the absence of oxygen) organic material is broken down in several steps by different types of microorganisms. The end-products are a gas containing mainly methane and carbon dioxide, referred to as biogas; and a slurry or solid fraction consisting of what is left of the treated substrate, referred to as digestate. Biogas can be produced from most types of organic raw material, except for lignin, which is not anaerobically degraded. The substrate composition will affect the yield of biogas and its content of methane (Tab. 1).

Landfill gas is produced during anaerobic digestion of organic materials in landfills and is very similar to biogas. Its methane content is generally lower than that of biogas, and landfill gas usually also contains nitrogen from air that seeps into the landfill gas during recovery. Landfill gas can also, in contrast to e.g. biogas from farms, contain a great number of trace gases.



Photo 1. Source separated household waste to be used as substrate for biogas production. The composition of biogas is depending on the type of substrates used in the digestion.

Tab. 1. Biogas yield and methane content of the biogas from different classes of substrates.

	Biogas yield (l/kg VS*)	Methane content (%)
Fat	1000–1250	70–75
Protein	600–700	68–73
Carbohydrate	700–800	50–55

*VS = Volatile Solids

There are different technologies for the biogas production, e.g. one stage, two stage and dry digestion. The substrate, the production technology and the collection of the gas, all affect the composition of the gas (Tab. 2).

To increase the quality of the raw biogas, the gas is usually cleaned of unwanted substances such as hydrogen sulphide, oxygen, nitrogen, water and particulates. The main reason for doing this is to prevent corrosion and mechanical wear of the equipment in which the biogas is used.

The main difference in the composition between biogas and natural gas relates to the carbon dioxide content. Carbon dioxide is one of the main components of biogas, while natural gas contains very low amounts. In addition, natural gas also contains higher levels of hydrocarbons other than methane. These differences result in a lower energy content of biogas per unit volume compared to natural gas (Tab. 2). By separating carbon dioxide from the biogas in an upgrading process, the energy content of upgraded biogas becomes comparable to natural gas.

Several countries have defined standards for grid injection of upgraded biogas or for utilization as vehicle

Tab. 2. Composition of biogas, landfill gas and natural gas.

	Biogas	Landfill gas	Natural gas (Danish)*	Natural gas (Dutch)
Methane (vol-%)	60–70	35–65	89	81
Other hydro carbons (vol-%)	0	0	9.4	3,5
Hydrogen (vol-%)	0	0-3	0	–
Carbon dioxide (vol-%)	30–40	15–50	0.67	1
Nitrogen (vol-%)	~0.2	5–40	0.28	14
Oxygen (vol-%)	0	0-5	0	0
Hydrogen sulphide (ppm)	0–4000	0–100	2.9	–
Ammonia (ppm)	~100	~5	0	–
Lower heating value (kWh/Nm ³)	6.5	4.4	11.0	8.8

*Average during 2007 (Energinet.dk).

Tab. 3. Selected standard requirements for grid injection or for utilization as vehicle fuel.

Compound	Unit	France		Germany		Sweden	Switzerland		Austria	The Netherlands
		L gas	H gas	L gas grid	H gas grid		Lim. inject.	Unlim. Inject		
Higher Wobbe index	MJ/Nm ³	42.48–46.8	48.24–56.52	37.8–46.8	46.1–56.5				47.7–56.5	43.46–44.41
Methane content	Vol-%					95–99	>50	>96		>80
Carbon dioxide	Vol-%	<2		<6			<6		≤2 ⁶	
Oxygene	Vol-%			<3			<0.5		≤0.5 ⁶	
	ppmV	<100								
	Mol%									<0.5
Hydrogen	Vol-%	<6		≤5			<5		≤4 ⁶	<12
CO ₂ +O ₂ +N ₂	Vol-%					<5				
Water dew point	°C	<-5 ¹		<t ⁴		<t ⁵ -5			<-8 ⁷	-10 ⁸
Relative humidity	p						<60 %			
Sulphur	mg/Nm ³	<100 ²		<30		<23	<30		≤5	<45
		<75 ³								

¹ At MOP (Maximal Operating Pressure) downstream from injection point

² Maximum permitted

³ Average content

⁴ Ground temperature

⁵ Ambient temperature

⁶ Mole percentage

⁷ At 40 bars

⁸ At 10 bars

fuel (Tab. 3). France, Germany and Switzerland have two levels of requirements for the upgraded biogas with different restrictions applied for the injection of low and high quality gas. Sweden has one standard that has been defined for biogas utilized as vehicle fuel.

In 2010 a mandate of the European Commission will be given to CEN to produce a set of biomethane standards for grid injection and and vehicle utilisation.

Cleaning of biogas

Apart from methane and carbon dioxide, biogas can also contain water, hydrogen sulphide, nitrogen, oxygen, ammonia, siloxanes and particles. The concentrations of these impurities are dependent on the composition of the substrate from which the gas was produced.

In those upgrading technologies where carbon dioxide is separated from the biogas, some of the other unwanted compounds are also separated. However, to prevent corrosion and mechanical wear of the upgrading equipment itself, it can be advantageous to clean the gas before the upgrading.

Removal of water

When leaving the digester, biogas is saturated with water vapour, and this water may condensate in gas pipelines and cause corrosion. Water can be removed by cooling, compression, absorption or adsorption. By increasing the pressure or decreasing the temperature, water will condensate from the biogas and can thereby be removed. Cooling can be simply achieved by burying the gas line equipped with a condensate trap in the soil. Water can also be removed by adsorption using e.g.



Photo 2. Digesters for industrial biogas production.

Photo 3. Part of natural gas grid into which upgraded biogas can be injected if it fulfils national standard and regulations.

Photo 4. Sign for natural gas grid.

SiO₂, activated carbon or molecular sieves. These materials are usually regenerated by e.g. heating or a decrease in pressure. Other technologies for water removal are absorption in glycol solutions or the use of hygroscopic salts.

Removal of hydrogen sulphide

Hydrogen sulphide is formed during microbiological reduction of sulphur containing compounds (sulphates, peptides, amino acids). The concentrations of hydrogen sulphide in the biogas can be decreased either by precipitation in the digester liquid or by treating the gas either in a stand alone vessel or while removing carbon dioxide.

Precipitation

Addition of Fe²⁺ ions or Fe³⁺ ions in the form of e.g. FeCl₂, FeCl₃ or FeSO₄, to the digester precipitates the almost insoluble iron sulphide that is removed together with the digestate. The method is primarily used in digesters with high sulphur concentration as a first measure or in cases where H₂S in the biogas is allowed to be high (e.g. higher than 1.000 ppm).

For the removal of H₂S from biogas, several technologies have been developed that will be described below.

Adsorption on activated carbon

Hydrogen sulphide is adsorbed on the inner surfaces of engineered activated carbon with defined pore sizes. Addition of oxygen (in the presence of water) oxidizes H₂S to plane sulphur that binds to the surface. In order

to increase the speed of the reaction and the total load, the activated carbon is either impregnated or doped (by addition of a reactive species before formation of the activated carbon) with permanganate or potassium iodide (KI), potassium carbonate (K₂CO₃) or zinc oxide (ZnO) as catalysers.

For grid injection or utilisation as vehicle fuel, only marginal amounts of O₂ are allowed in the gas. Hence oxidation of the sulphur is not suitable. In those cases mostly KI-doped carbon or permanganate impregnated carbon is used because addition of oxygen is not required in the case of KI under reduced loading.

While ZnO impregnated carbon is rather expensive, H₂S removal is extremely efficient with resulting concentrations of less than 1ppm.

Chemical Absorption

One of the oldest methods of H₂S removal involves sodium hydroxide (NaOH) washing. Because of the high technical requirement to deal with the caustic solution, it's application is hardly applied anymore except when very large gas volumes are treated or high concentrations of H₂S are present.

Hydrogen sulphide can also be adsorbed using iron oxide-coated (Fe(OH)₃ or Fe₂O₃) support material (mostly pressed minerals, sometimes wood chips). In this treatment biogas is passed through iron oxide-coated material. Regeneration is possible for a limited number of times (until the surface is covered with natural sulphur), after which the tower filling has to be renewed. The process operates with two columns, one is absorbing, while the other is re-oxidized.

If a small amount of air is present in the biogas, the system can operate with one column but loading is limited when compared to the two-column system. This method has been used worldwide in sewage sludge treatment plants, before Fe³⁺ addition became standard for the simultaneous removal of phosphate.

Iron oxide is also the desulphurizing agent in SOXSIA® (Sulphur Oxidation and Siloxanes Adsorption), a catalyst developed by Gastreatment Services B.V. SOXSIA® that adsorbs siloxanes and removes H₂S from the raw gas. Up to 2000 ppm of H₂S can be removed from the



Photo 5. Column for hydrogen sulphide removal with activated carbon. After desulphurisation the gas enters a PSA-upgrading plant.

gas at 40°C, atmospheric pressure and with a capacity of 1000 Nm³ raw gas/hour. Another example of a product commercially available for adsorption of hydrogen sulphide from biogas is Sulfa Treat®.

Hydrogen sulphide can be absorbed in e.g. a ferric chelate solution in which Fe³⁺ ions are reduced to Fe²⁺ ions while hydrogen sulphide is oxidized to elementary sulphur. The ferric chelate solution is regenerated in a second vessel by addition of oxygen and water. Chelate technologies are designed for high loads and are usually not applied in biogas plants.

In a process recently developed by Procede, a Dutch company, hydrogen sulphide removal is based on the precipitation reaction between hydrogen sulphide and a metal ion in an aqueous solution. The metal sulphide that is formed precipitates almost immediately. The metal ion is regenerated by using oxygen which converts the bound sulphur to sulphur dioxide that can be used to produce sulphuric acid or gypsum. The process is able to clean biogas down to less than 1 ppm hydrogen sulphide. It has so far been tested in a pilot plant (5 Nm³/h), and will be available for biogas plants up to 1500 Nm³/h.

Acrion Technologies Inc. has developed a system called CO₂ Wash® for the cleaning of landfill gas. The CO₂ Wash® removes siloxanes, sulphur compounds, halogenated compounds and NMHC (non-methane hydrocarbons) from landfill gas. The unwanted compounds are separated by liquid carbon dioxide originating from

the landfill gas (Fig. 3). The removed compounds dissolved in the liquid carbon dioxide can be incinerated together with landfill gas. Other streams from the CO₂ Wash® are a pure liquid carbon dioxide stream and a gas stream containing methane and car-

bon dioxide. The liquid carbon dioxide is 99.99% pure and the concentrations of siloxanes, chlorinated hydrocarbons and sulphur compounds in the methane and carbon dioxide stream are all below the detection limits of 5 ppb, 10 ppb and 100 ppb, respectively, according to Acrion.

Biological treatment

Hydrogen sulphide can be oxidized by microorganisms of the species *Thiobacillus* and *Sulfolobus*. The degradation requires oxygen and therefore a small amount of air (or pure oxygen if levels of nitrogen should be minimized) is added for biological desulphurization to take place. The degradation can occur inside the digester and can be facilitated by immobilizing the microorganisms occurring naturally in the digestate. An alternative is to use a trickling filter which the biogas passes through when leaving the digester. In the trickling filter the microorganisms grow on a packing material. Biogas with added air meets a counter flow of water containing nutrients. The sulphur containing solution is removed and replaced when the pH drops below a certain level. Both methods are widely applied, however they are not suitable when the biogas is used as vehicle fuel or for grid injection due to the remaining traces of oxygen. An alternative system has been developed by Profactor, where the absorption of the H₂S is separated from the biological oxidation to sulphur. Hence, the biogas flow remains free of oxygen.

Removal of oxygen and nitrogen

Oxygen is not normally present in biogas since it should be consumed by the facultative aerobic microorganisms in the digester. However, if there is air present in the digester nitrogen will still be present in the gas when leaving the digester. Oxygen and nitrogen can be present in landfill gas if the gas is collected using an under pressure. These gases can be removed by adsorption with activated carbon, molecular sieves or membranes. They can also to some extent be removed in desulphurisation processes or in some of the biogas upgrading processes. Both compounds are difficult (i.e. expensive) to remove hence, their presence should be avoided unless the biogas is used for CHPs or boilers.

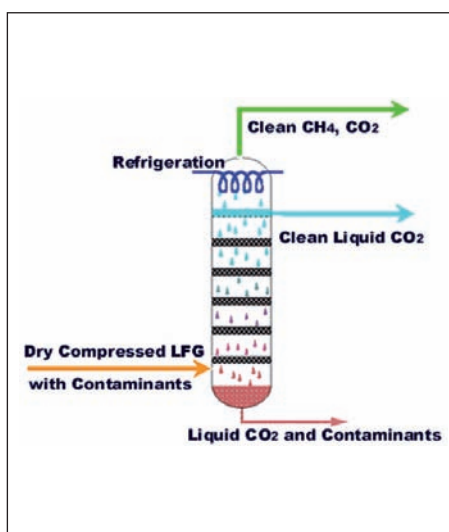


Fig. 3. Schematic view of the CO₂ Wash® (Acrion).

Removal of ammonia

Ammonia is formed during the degradation of proteins. The amounts that are present in the gas are dependent upon the substrate composition and the pH in the digester. Ammonia is usually separated when the gas is dried or when it is upgraded. A separate cleaning step is therefore usually not necessary.

Removal of siloxanes

Siloxanes are compounds containing a silicon-oxygen bond. They are used in products such as deodorants and shampoos, and can therefore be found in biogas from sewage sludge treatment plants and in landfill gas. When siloxanes are burned, silicon oxide, a white powder, is formed which can create a problem in gas engines. Siloxanes can be removed by cooling the gas, by adsorption on activated carbon (spent after use), activated aluminium or silica gel, or by absorption in liquid mixtures of hydrocarbons. Siloxanes can also be removed whilst separating hydrogen sulphide, as described under "Removal of hydrogen sulphide".

Removal of water

Biogas is saturated with water vapour when it leaves the digester. The water can condensate in gas pipelines and together with sulphur oxides may cause corrosion. By increasing the pressure or decreasing the temperature, water will condense from the biogas and can thereby be removed. Cooling can be achieved naturally by leading it through a pipe in the soil equipped with a condensate trap or with an electric cooler. Water can also be removed by adsorption using SiO_2 , activated charcoal or molecular sieves. These materials are usually regenerated by heating and/or a decrease in pressure. Other technologies for water removal are absorption in glycol solutions or the use of hygroscopic salts.

Removal of particulates

Particulates can be present in biogas and landfill gas and can cause mechanical wear in gas engines and gas turbines. Particulates that are present in the biogas are separated by mechanical filters.

Full scale technology for biogas upgrading

In the present report upgrading of biogas or landfill gas is defined as removal of carbon dioxide from the gas. This will result in an increased energy density since the concentration of methane is increased. Several technologies for biogas upgrading are commercially available and others are at the pilot or demonstration plant level. Some of these technologies are described below and reviewed in terms of recent developments. A list of suppliers of biogas upgrading plants can be found at the end of this brochure.

Pressure Swing Adsorption (PSA)

With this technique, carbon dioxide is separated from the biogas by adsorption on a surface under elevated pressure. The adsorbing material, usually activated carbon or zeolites, is regenerated by a sequential decrease in pressure before the column is reloaded again, hence the name of the technique. An upgrading plant, using this technique, has four, six or nine vessels working in parallel. When the adsorbing material in one vessel becomes saturated the raw gas flow is switched to another vessel in which the adsorbing material has been regenerated. During regeneration the pressure is decreased in several steps. The gas that is desorbed during the first and eventually the second pressure drop may be returned to the inlet of the raw gas, since it will contain some methane that was adsorbed together with carbon dioxide. The gas desorbed in the following pressure reduction step is either led to the next column or if it is almost entirely methane free it is released to the atmosphere.

If hydrogen sulphide is present in the raw gas, it will be irreversibly adsorbed on the adsorbing material. In addition, water present in the raw gas can destroy the structure of the material. Therefore hydrogen sulphide and water needs to be removed before the PSA-column.

A Canadian company, Quest Air, has developed a PSA system for cleaning and upgrading of hydrogen. Acron Systems, a Swiss company, has further developed the

system for the upgrading of biogas. The system consists of nine adsorption columns oriented in a circle and gas distribution between them is controlled by a single valve. A small amount of the purified methane stream is used to purge each vessel and thus prepare it for a new cycle. The simple construction results in a product of compact size which provides for low capital and installations costs, for small scale installations (up to 250 Nm³/h).

Absorption

In an upgrading plant using the absorption technique the raw biogas meets a counter flow of liquid in a column which is filled with plastic packing (in order to increase the contact area between the gas and the liquid phase). The principal behind the absorption technique is that carbon dioxide is more soluble than methane. The liquid leaving the column will thus contain increased concentration of carbon dioxide, while the gas leaving the column will have an increased concentration of methane. Three examples of the absorption technology using different types of absorbents are water scrubbing, organic physical scrubbing and chemical scrubbing.

Water scrubbing

Carbon dioxide has a higher solubility in water than methane. Carbon dioxide will therefore be dissolved to a higher extent than methane, particularly at lower temperatures (Fig. 4).

In the scrubber column carbon dioxide is dissolved in the water, while the methane concentration in the gas phase increases. The gas leaving the scrubber has therefore an increased concentration of methane. The water leaving the absorption column is transferred to a flash

tank where the dissolved gas, which contains some methane but mainly carbon dioxide, is released and transferred back to the raw gas inlet. If the water should be recycled it is transferred to a desorption column filled with plastic packing, where it meets a counter flow of air, into which carbon dioxide will be released. The water is cooled down to achieve the large difference in solubility between methane and carbon dioxide before it is recycled back to the absorption column.

Water scrubbing is the most common upgrading technique and plants are commercially available from several suppliers in a broad range of capacities. Biorega AB has developed a water scrubber that is designed for small raw gas flows. In Biorega's system the carbon dioxide is desorbed by a vacuum pump connected to the desorption column. In 2004, a pilot plant was built with a capacity of treating 12 Nm³ raw biogas per hour. A second demonstration plant with a capacity of 15–18 Nm³/h is now being built.

An alternative water scrubber technology has been developed by Metener, a Finnish company. In their process biogas is upgraded and pressurized (to around 150 bars) simultaneously in batch mode. Raw biogas is compressed and added to a column. The column is then filled with water using high pressure water pumps. Carbon dioxide and sulphurous compounds are dissolved in the water. After the scrubbing clean pressurized gas leaves the column and the water is regenerated in a flash tank followed by a regeneration tank. The system consists of two water scrubbing columns working in parallel in different phases (while one is being filled the other one is being emptied). The technology is most suitable for biogas flows of 30–100 Nm³/h and has been demonstrated in a pilot plant with a capacity of 40 Nm³/h in Laukaa, Finland.



Photo 6. Upgrading plant, Malmö Sweden, using the PSA technology of Carbotech

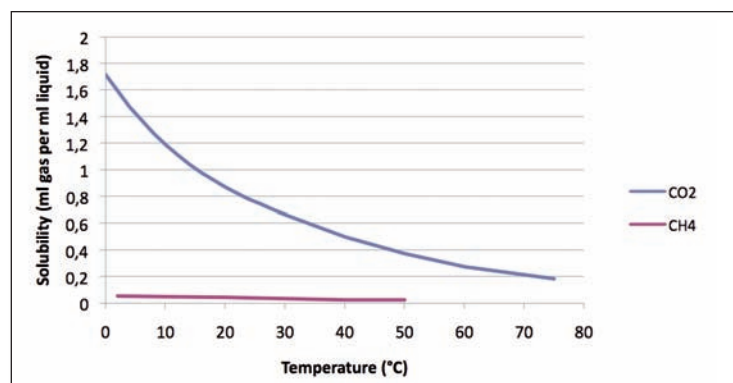


Fig. 4. Solubility of methane and carbon dioxide in water (Data source: Gas Encyclopaedia).



Photo 7. Kristianstad, Sweden, upgrading plant with a water scrubber. In the back the absorption column, flash tank and desorption column can be seen.

Organic physical scrubbing

Organic physical scrubbing is very similar to water scrubbing, with the important difference that the carbon dioxide is absorbed in an organic solvent such as polyethylene glycol. Carbon dioxide is more soluble in polyethylene glycol than in water and for the same upgrading capacity the flow of the liquid phase can be lower and so the plant can be smaller. The polyethylene glycol solution is regenerated by heating and/or depressurizing. Hydrogen sulphide, water, oxygen and nitrogen may be removed together with carbon dioxide. However, more often they are removed prior to upgrading. Selexol® and Genosorb® are examples of trade names for liquids used in organic physical scrubbing.

Chemical scrubbing

Chemical scrubbers use amine solutions. Carbon dioxide is not only absorbed in the liquid, but also reacts chemically with the amine in the liquid. Since the chemical reaction is strongly selective, the methane loss might be as low as <math><0.1\%</math>. Part of the liquid is lost due to evaporation, and has to be replaced. The liquid in which carbon dioxide is chemically bound is regenerated by heating. Two types of compounds are used: mono ethanol amine (MEA) and di-methyl ethanol amine (DMEA).

If hydrogen sulphide is present in the raw gas, it will be absorbed in the amine scrubber solution and higher temperatures will be needed for the regeneration. Therefore it is advisable to remove it before absorption in the amine scrubber.

Membranes

Dry membranes for biogas upgrading are made of materials that are permeable to carbon dioxide, water and ammonia. Hydrogen sulphide, and oxygen permeate through the membrane to some extent while nitrogen and methane only pass to a very low extent. Usually membranes are in the form of hollow fibres bundled together. The process is often performed in two stages. Before the gas enters the hollow fibres it passes through a filter that retains water and oil droplets and aerosols, which would otherwise negatively affect the membrane performance. Additionally, hydrogen sulphide is usually removed by cleaning with activated carbon before the membrane.

Membrane separation is one of the classical methods for landfill gas upgrading. The first plants were built in the late 1970's in the US and later in the Netherlands. However, the early designs operating at elevated pressures (up to 30 bars) suffered from considerable methane losses (up to 25%). Newer designs operate around 8 bars with far lower methane losses.

In 2007 a plant using a new design developed conjointly by the Technical University of Vienna and the company Axiom was commissioned for an installation in Bruck, Austria. Its capacity is 180 Nm³/h and the gas is upgraded in a two-stage process minimizing the methane content in the off-gas. After upgrading the gas is injected into the gas grid.

In a demonstration project in Ohio, landfill gas, which was first purified with the CO₂ Wash® process (see description under "Removal of hydrogen sulphide") was then upgraded with membrane technology after which the upgraded gas was liquefied using liquid nitrogen.



Photo 8. Upgrading plant in Gothenburg, Sweden, using the chemical scrubber technology. To the left the stripper column where the amine solution is regenerated by heat and to the right the absorption column.

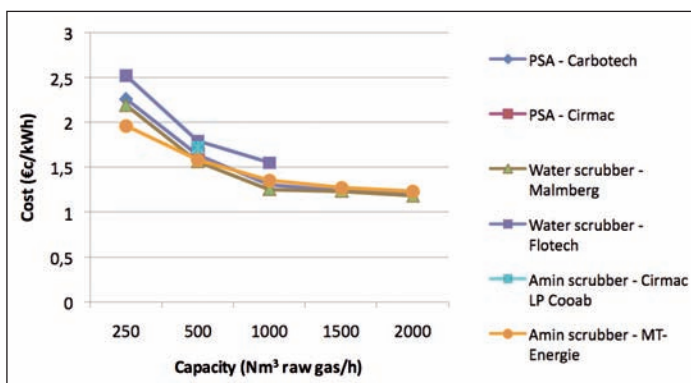
The concentration of carbon dioxide was decreased from 30% to 50 ppm by the two membranes working in series (Acrion). By cooling the methane rich gas after the membranes, nitrogen can be separated from methane, due to the difference in their boiling points (Tab. 4).

Comparison of different upgrading techniques

The most widely used technologies for biogas upgrading are pressure swing adsorption, water scrubbing, organic physical scrubbing and chemical scrubbing. Their characteristics as given by the technology providers are summarized in Tab. 5. However, it is important to remember that the best technology to choose is based on specific parameters at the plant, such as the availability of cheap heat and the electricity price. It should also be noted that it is often possible to lower the methane loss, but at the expense of a higher energy consumption. Methane that is lost in the upgrading process can be prevented from causing a methane slip to the atmosphere using technologies reviewed under “Removal of methane from the off-gas”.

Today, technological developments have led to cheaper and more efficient plants thanks to the increasing interest in upgrading biogas. The demand for more plants has also led to the development of standardized upgrading units which also decreases the costs.

The upgrading costs of established techniques are dependent on the specific technology, but most importantly on the size of the plant (Fig. 5). However, the field of biogas upgrading is developing rapidly and thus the cost development would also be expected to change. Today, there are commercially available plants for capacities lower than 250 Nm³/h, while also plants larger than 2000 Nm³/h are being built. These developments and also the fact that more plants are being built will likely lead to lower prices.



Tab. 4. Boiling points at atmospheric pressure (Gas Encyclopaedia).

Compound	Boiling point (K)	Boiling point (°C)
CO ₂	194.65*	-78.50*
Methane	111.63	-161.52
Oxygene	90.18	-182.97
Nitrogen	77.347	-195.803

* Sublimation point

New developments in upgrading technology

Apart from the continuous improvements of the established technologies, new technologies are also being developed. Some of them will be highlighted below. However, it should be emphasized that the stage of development might be quite different.

Cryogenic upgrading

Cryogenic upgrading makes use of the distinct boiling/sublimation points of the different gases particularly for the separation of carbon dioxide and methane. The raw biogas is cooled down to the temperatures where the carbon dioxide in the gas condenses or sublimates and can be separated as a liquid or a solid fraction, while methane accumulates in the gas phase. Water and siloxanes are also removed during cooling of the gas.

The sublimation point of pure carbon dioxide is 194.65 K (Fig. 6). However, the content of methane in the biogas affects the characteristics of the gas, i.e. higher pressures and/or lower temperatures are needed to condense or sublimate carbon dioxide when it is in a mixture with methane.

Cooling usually takes place in several steps in order to remove the different gases in the biogas individually and to optimize the energy recovery. In the GPP[®] system (Fig. 7) from Gastreatment Services B.V. biogas is first compressed to 17–26 bar and then cooled to -25°C. In this step water, hydrogen sulphide, sulphur dioxide, halogens and siloxanes are removed from the gas. The gas is then led through a coalescence filter and then through a SOXSIA[®] catalyst which removes any remaining contaminants. Carbon dioxide is removed in two

Fig. 5. Estimated cost of biogas upgrading plants using different technologies (Urban et al.2008).

Tab.5. Comparison between selected parameters for common upgrading processes (Urban et al. 2008).

Parameter	PSA	Water scrubbing	Organic physical scrubbing	Chemical scrubbing
Pre-cleaning needed ^a	Yes	No	No	Yes
Working pressure (bar)	4–7	4–7	4–7	No pressure
Methane loss ^b	<3% / 6–10% ^f	<1% / <2% ^g	2–4%	<0.1%
Methane content in upgraded gas ^c	>96%	>97%	>96%	>99%
Electricity consumption ^d (kWh/Nm ³)	0.25	<0.25	0.24–0.33	<0.15
Heat requirement (°C)	No	No	55–80	160
Controllability compared to nominal load	+/- 10–15%	50–100%	10–100%	50–100%
References ^e	>20	>20	2	3

^a Refers to raw biogas with less than 500 mg/m³ of H₂S. For higher concentrations, pre-cleaning is recommended also for the other techniques.

^b The methane loss is dependent on operating conditions. The figures given here refer to figures guaranteed by the manufacturer or provided by operators.

^c The quality of biomethane is a function of operational parameters. Figures given refer to figures guaranteed by the manufacturer or provided by operators, based on air-free biogas.

^d Given in kWh/Nm³ of raw biogas, compressed to 7 bar(g).

^e Number of references reviewed. Some are pilot plants.

^f <3% CarboTech, / 6–10% QuestAir.

^g <1% Malmberg / <2% Flotech.

further stages. In the first stage the gas is cooled down to between -50°C and -59°C where 30-40% of the carbon dioxide is removed as a liquid. In the second stage the remaining carbon dioxide is removed as a solid. Since the carbon dioxide is solid at this stage the process needs a second column, which is used while defrosting and removing carbon dioxide from the first column. Gastreatment Services B.V. is developing the GPP[®]plus system which in addition to upgrading biogas will produce liquid methane as an end-product. This system is in the research phase and a pilot plant has been in operation in the Netherlands since the beginning of 2009. By decreasing the temperature enough to produce liquid methane, it is also possible to separate nitrogen, which is an advantage when upgrading landfill gas.

Prometheus has a technique for upgrading landfill gas which consists of the following three steps:

1. *Pre-Purification Module:* Corrosive sulphur compounds, low concentrations of non-methane-organic compounds (including siloxanes) and water are removed and the gas is compressed to around 4.4 bars.
2. *Bulk Purification Module:* Carbon dioxide is removed by freezing it while simultaneously pre-cooling methane and nitrogen.
3. *Liquefaction and Post-Purification Module:* The purified gas is liquefied and the concentration of methane is increased by dynamic flash evaporation of the nitrogen.

A pilot-scale plant using this technology was designed in 2000 in Victoria, B.C. Canada. Liquid methane



Photo 9 and 10. Upgraded biogas can be used as a vehicle fuel in cars and buses.

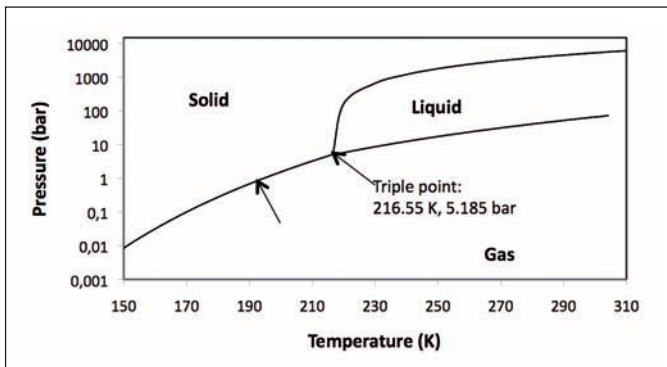


Fig. 6. Phase diagram for carbon dioxide (Data source: Gas Encyclopaedia).

was produced with a purity of 96%. The first commercial scale plant was built in 2006 at the Bowerman Landfill, CA, USA, in a cooperation between Prometheus and Montauk Energy Capital. The plant is designed for production of 19 m³ of liquid methane per day. There are plans to expand the production to 150 m³ of liquid methane per day in the future.

In situ methane enrichment

Carbon dioxide is to some extent soluble in water and therefore some carbon dioxide will be dissolved in the liquid phase of the digester tank (Fig. 8). In upgrading with the in situ methane enrichment process, sludge from the digester is circulated to a desorption column and then back to the digester. In the desorption column carbon dioxide is desorbed by pumping air through the sludge (Fig. 8). The constant removal of carbon dioxide from the sludge leads to an increased concentration of methane in the biogas phase leaving the digester (Lindberg, 2003).

Process simulations have shown that it may be possible to reach a biogas quality of 95% methane with a methane loss below 2%. Cost estimations have shown that for a raw gas flow of below 100 Nm³/h, the cost can

be one third of the cost of conventional techniques. A pilot plant with a digester volume of 15 m³ and a 140 dm³ bubble column has been constructed and tested (Nordberg et al. 2006). In-situ methane enrichment will change the buffer capacity of the sludge, but results of the same study showed that desorption with air did not have a negative effect on the methane yield in the digester. In experiments where different sludge and air flows were tested the highest methane content obtained was 87% with 2% nitrogen and a methane loss of 8% in the off-gas from the desorption column (Nordberg et al. 2006). The technology is relatively simple and there is no need for much auxiliary equipment such as pressurized tanks. Therefore it has a potential for a lower upgrading cost compared to other techniques. However, the process is limited to smaller plants where a high methane concentration (>95%) is not needed. It is primarily suited for sludge that is easy to pump. If the technique is applied to a digester using fibrous substrates, the concentration of nitrogen might increase due to air bubbles attaching to the material when it passes through the desorption column.

Ecological lung

The enzyme carboanhydrase is present in our blood where it catalyses the dissolution of carbon dioxide that is formed during metabolism in our cells. The enzyme catalyses the reaction:



The dissolved carbon dioxide, in the form of carbonate, is then transported to our lungs where the same enzyme catalyses the reverse reaction where carbon dioxide and water is formed.

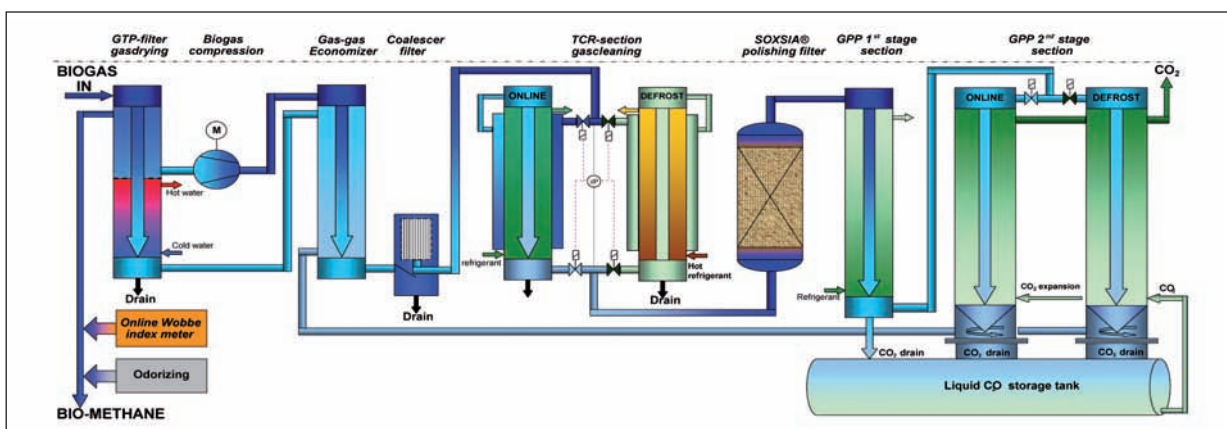


Fig. 7. Schematic view of GPP® system (Gastreatment Services).

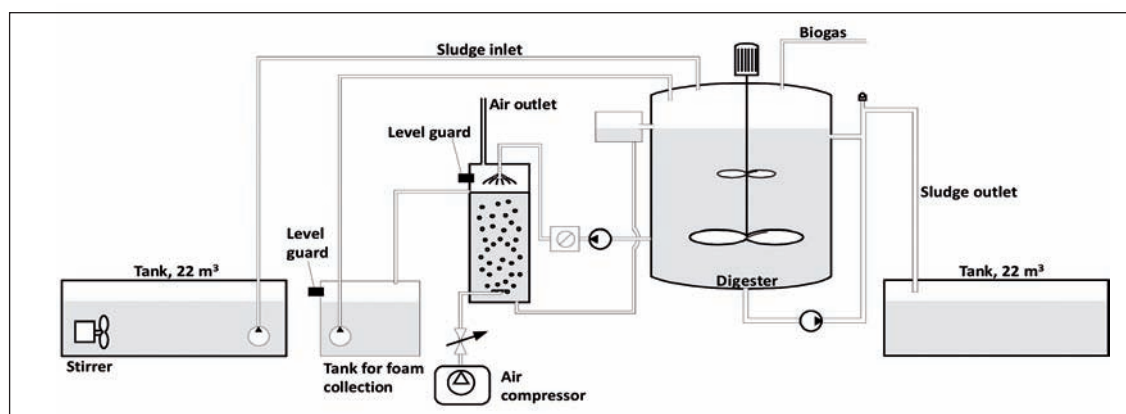


Fig. 8. Schematic view of in-situ methane enrichment research plant. (Courtesy of Åke Nordberg, SLU, Sweden).

The enzyme can also be used to dissolve carbon dioxide from biogas and thereby remove it from the gas. The production cost of the enzyme is still high and the viability of the process is affected by factors such as the lifetime of immobilized enzyme. A research group in Lund, Sweden, has studied the use of carboanhydrase for biogas upgrading and in one project the enzyme was produced with an addition of 6 histadines that were used to attach the enzyme to a solid carrier (Mattiasson, 2005). In the same study it was shown that biogas can be purified up to a methane content of 99%. CO₂ Solution Inc. is a Canadian company that has developed this technique and has a patent for a bioreactor using the enzyme for dissolving carbon dioxide. They do not only focus on biogas upgrading, but are also looking at this technique for e.g. ventilation. They are currently focusing their research projects on enzyme immobilization, bioreactor mechanics, enzyme cloning and production and technology applications (CO₂ Solution Inc.).

Removal of methane from the off-gas

It is important to minimize the loss of methane in order to achieve an economical viable upgrading plant. However, it is also important to minimize the methane slip since methane is a strong greenhouse gas. Thus the release of methane to the atmosphere should be minimized by treating the off-gas, air or water streams leaving the plant even though the methane cannot be utilized. Methane can be present in the off-gas leaving a PSA-column, in air from a water scrubber with water recirculation or in water in a water scrubber without water recirculation.

The off-gas from an upgrading plant seldom contains a high enough methane concentration to maintain a flame without addition of natural gas or biogas. One way of limiting the methane slip is to mix the off-gas with air that is used for combustion. Alternatively the methane can be oxidized by thermal or catalytic oxidation.

The VOCSIDIZER from Megtec is an example of a device designed for regenerative thermal oxidation. The VOCSIDIZER consists of a heat transfer bed filled with ceramic media. When the off-gas moves through the porous media it is heated until it is hot enough for methane oxidation with oxygen to form water vapour and carbon dioxide. The temperature of the gas leaving the VOCSIDIZER will tend to increase due to a shift of the temperature profile towards the end of the heat transfer bed, and therefore the flow of off-gas is periodically reversed. Once running, the heat generated by the oxidation is enough to maintain the function of the VOCSIDIZER.

Another example of a commercially available thermal oxidation device is the FLOX (Flameless Oxidation), such as the FLOX[®]-LCV from e-flox GmbH. In the first step the oxidation chamber is heated up to 650°C by using natural gas (or biogas). Once this temperature is reached, the off-gas is preheated to 450°C by the exhaust gas. The remaining difference from the oxidation temperature of 950°C can be delivered by the off-gas during the oxidation. The surplus heat in the exhaust gas after pre-heating of the off-gas is recovered with a heat exchanger and used for heating purposes.

Another option is to catalytically oxidize the methane. In this case, the oxidation takes place at the surface of the catalyst. The catalyst lowers the energy needed to oxidize the methane, thus enabling oxidation at a lower temperature. The active component of the catalyst is platinum, palladium or cobalt.

List of upgrading plants

COUNTRY	PLACE	SUBSTRATE	UTILISATION	CH ₄ REQUIREMENTS (%)	TECHNOLOGY	PLANT CAPACITY (NM ³ /H RAW GAS)	IN OPERATION SINCE
Austria	Bruck/Leitha	Biowaste	Gas grid	97	Membrane	180	2007
	Linz	Sewage	Gas grid	97	Water scrubber	800	2008
	Margarethen am Moos	Energy Crops & Manure	Vehicle fuel	>95	Membrane	70	2007
	Pucking	Manure	Gas grid	97	PSA	10	2005
	Reitbach / Eugendorf	Energy crops	Gas grid Vehicle fuel	97	PSA	150	2008
Canada	Berthierville, (QC)	Landfill gas	Gas grid		Membrane		2003
France	Lille	Biowaste	Vehicle fuel	97	Waterscrubber	2*600	2007
	Lille Marquette				Water scrubber	100	2009
Germany	Altenstadt	Biowaste	Gas grid		Water scrubber	1250	2009
	Bottrop	Sewage sludge	Vehicle fuel		PSA	120	2008
	Burgrieden	Energy crops	Gas grid		PSA	300	2008
	Einbeck	Energy crops	Gas grid		Chemical scrubber	1000	2009
	Ettlingen	Energy crops	Gas grid		PSA	600	2008
	Forchheim	Energy crops	Gas grid		Genosorb scrubber	1000	2009
	Gemeinde Graben Landkreis Augsburg	Energy crops	Gas grid		PSA	1000	2008
	Godenstedt	Energy crops	Gas grid		Chemical scrubber	600	2009
	Güstrow, M-V	Energy crops	Gas grid		Water scrubber	10000	2009
	Hardegesen	Energy crops	Gas grid		Chemical scrubber	1000	2009
	Horn-Bad Meinberg (NRW)	Energy crops	Gas grid		Chemical scrubber	2000	2009
	Jameln	Manure Energy crops	Vehicle fuel, gas grid		Genosorb scrubber	160	2005
	Kerpen	Energy crops	Gas grid		PSA	1000	2008
	Ketzin	Energy crops	Gas grid		PSA	400	2008
	Könnern I	Manure, energy crops	Gas grid		Water scrubber	1250	2007
	Könnern II	Energy crops	Gas grid		Chemical scrubber	3400	2009
	Lüchow	Energy crops	Gas grid		Water scrubber	1250	2008
	Maihingen	Energy crops	Gas grid		Water scrubber	1250	2007
	Mühlacker	Energy crops	Gas grid		PSA	920	2007
	Niederndodeleben	Energy crops	Gas grid		Water scrubber	1250	2008
	Pliening	Energy crops	Gas grid		PSA	920	2006
	Rathenow	Energy crops	Gas grid		Genosorb scrubber	1130	2009
	Ronnenberg	Energy crops	Gas grid		Genosorb scrubber	650	2008
	Schwandorf I	Energy crops	Gas grid		Organic scrubber	1000	2007
	Schwandorf II	Energy crops	Gas grid		PSA	2000	2008
	Straelen	Energy crops, manure	Gas grid		PSA	1000	2006
	Utzensdorf	Biowaste	Gas grid	96	PSA	100	2009
	Werlte	Manure, biowaste	Gas grid		PSA	500	2007
Wixhausen (Darmstadt)	Manure, energy crops	Gas grid		Waterscrubber	300	2008	
Wriezen	Energy crops	Gas grid		PSA	1200	2009	
Wüsting	Energy crops	Gas grid		PSA	1200	2009	
Iceland	Reykjavik	Landfill gas	Vehicle fuel		Water scrubber	700	2005
Japan	Kobe	Sewage sludge	Vehicle fuel	97	Water scrubber	100	2004
	Kobe	Sewage sludge	Vehicle fuel	97	Water scrubber	2*225	2007
Norway	Fredrikstad		Vehicle fuel		PSA		2001
	Oslo	Sewage sludge	Vehicle fuel		Chemical Scrubber	750	2009
	Stavanger	Sewage sludge, biowaste	Gas grid		Chemical scrubber	500	2009
The Netherlands (information kindly supplied by Erik Polman, Kiwa)	Beverwijk	Landfill gas	Gas grid	88	Membrane		2006
	Collendoorn	Landfill gas	Gas grid	88	Membrane	375	1993
	Mijdrecht	Sewage sludge	Gas grid	88			2009
	Nuenen	Landfill gas	Gas grid	88	PSA	1500	1990
	Tilburg-De Spinder	Landfill gas	Gas grid	88	Water scrubber	2100	1987
Wijster	Landfill gas	Gas grid	88	PSA	1150	1989	
South Korea	Seoul				Waterscrubber	150	2008
Spain	Madrid	Biowaste	Vehicle fuel	96.5	Water scrubber	4000	2008
	Vacarisses (Barcelona)	Landfill gas	Vehicle fuel	>85	Chemical scrubber	100	2005

List of upgrading plants

Biogas upgrading

COUNTRY	PLACE	SUBSTRATE	UTILISATION	CH ₄ REQUIREMENTS (%)	TECHNOLOGY	PLANT CAPACITY (NM ³ /H RAW GAS)	IN OPERATION SINCE
Sweden	Bjuv	Biowaste, manure	Gas grid	97	PSA	500	2007
	Boden	Sewage sludge, biowaste	Vehicle fuel	97	Water scrubber	360	2007
	Borås	Biowaste, sewage sludge	Vehicle fuel	97	Chemical scrubber	450	2002
	Bromma, Stockholm	Sewage sludge	Vehicle fuel	97	PSA	250	2002
	Bromma, Stockholm	Sewage sludge	Vehicle fuel	97	PSA	250	2003
	Eskiltuna	Biowaste, sewage sludge	Vehicle fuel	97	Water scrubber	330	2003
	Eslöv	Biowaste, sewage sludge	Vehicle fuel	97	Water scrubber	80	1999
	Falkenberg	Sewage sludge, biowaste, energy crops	Gas grid	97	Chemical scrubber	750	2009
	Falköping	Sewage sludge	Vehicle fuel	97	Water scrubber	200	2007
	Göteborg	Sewage sludge, biowaste	Gas grid	97	Chemical scrubber	1600	2007
	Helsingborg	Biowaste, manure	Vehicle fuel and Gas grid	97	PSA	350	2001
	Helsingborg	Biowaste, manure	Vehicle fuel and Gas grid	97	Water scrubber	650	2007
	Helsingborg	Sewage sludge	Gas grid	97	Water scrubber	250	2007
	Henriksdal, Stockholm	Sewage sludge	Vehicle fuel	97	Water scrubber	600	2004
	Henriksdal, Stockholm	Sewage sludge	Vehicle fuel	97	Water scrubber	800	2006
	Himmerfjärden, Stockholm	Sewage sludge	Vehicle fuel	97	Chemical scrubber	800	2009
	Jönköping	Sewage sludge, biowaste	Vehicle fuel	97	Water scrubber	300	2000
	Kalmar	Sewage sludge, manure	Vehicle fuel	97	Chemical scrubber	200	2008
	Katrineholm	Sewage sludge	Vehicle fuel	97	Water scrubber	80	2009
	Kristianstad	Biowaste, manure, sewage sludge	Vehicle fuel	97	Water scrubber	280	1999
	Kristianstad	Biowaste, manure, sewage sludge	Vehicle fuel	97	Water scrubber	600	2006
	Laholm	Biowaste, manure	Gas grid	97	Water scrubber	500	2000
	Linköping	Sewage sludge, biowaste	Vehicle fuel	97	Water scrubber	2*330	1997
	Linköping	Sewage sludge, biowaste	Vehicle fuel	97	Water scrubber	1400	2002
	Malmö	Sewage sludge	Gas grid	97	PSA	500	2008
	Motala	Sewage sludge	Vehicle gas	97	Water scrubber	80	2009
	Norrköping	Sewage sludge	Vehicle fuel	97	Water scrubber	250	2004
	Norrköping	Distiller's waste, Energy crops	Vehicle fuel	97	Water scrubber	240	2006
	Skellefteå	Sewage sludge	Vehicle fuel	97	Water scrubber	250	2005
	Skövde	Sewage sludge, slaughter waste	Vehicle fuel	97	PSA	140	2002
	Trollhättan	Biowaste, sewage sludge	Vehicle fuel	97	Water scrubber	200	1995
	Trollhättan	Biowaste, sewage sludge	Vehicle fuel	97	Water scrubber	400	2001
	Ulricehamn	Sewage sludge	Vehicle fuel	97	PSA	20	2003
	Uppsala	Sewage sludge, biowaste	Vehicle fuel	97	Water scrubber	400	2001
Västervik	Sewage sludge	Vehicle fuel	97	Water scrubber	130	2009	
Västerås	Biowaste, sewage sludge	Vehicle fuel	97	Water scrubber	650	2004	
Örebro	Sewage sludge	Vehicle fuel	97	Water scrubber	450	2007	
Örebro	Sewage sludge	Vehicle fuel	97	Water scrubber	2000	2009	
Östersund	Sewage sludge	Vehicle fuel	97	Water scrubber	200	2006	
Switzerland	Bachenbülach	Biowaste	Gas grid and vehicle gas	96	PSA	50	1996
	Berne	Sewage sludge	Gas grid	96	PSA	300	2007
	Bischofszell	Sewage sludge	Gas grid	96	Genosorb scrubber	100	2007
	Jona	Biowaste	Gas grid	96	Genosorb scrubber	55	2005
	Inwil	Biowaste, manure	Gas grid	96	PSA	225	2009
	Lavigny	Biowaste	Gas grid	96	PSA	150	2009
	Lucerne	Sewage sludge	Gas grid	96	PSA	75	2004
	Obermeilen	Sewage sludge	Gas grid	96	Chemical scrubber	100	2008
	Otelfingen	Biowaste	Vehicle gas	96	PSA	50	1998
	Pratteln	Biowaste	Gas grid	96	Genosorb scrubber	300	2006
Roche	Sewage sludge	Gas grid	96	PSA	250	2008	

An updated version of the list of reference plants can be found at www.iea-biogas.net.

COUNTRY	PLACE	SUBSTRATE	UTILISATION	CH ₄ REQUIREMENTS (%)	TECHNOLOGY	PLANT CAPACITY (NM ³ /H RAW GAS)	IN OPERATION SINCE
Switzerland	Romanshorn	Sewage sludge	Gas grid		Genosorb scrubber	100	2007
	Rümlang	Biowaste	Vehicle fuel	96	PSA	30	1995
	Samstagern	Biowaste	Gas grid	96	PSA	50	1998
	Utzensdorf	Biowaste	Gas grid	96	PSA	100	2009
	Widnau	Agricultural co-digestion	Gas grid	96	PSA	100	2007
USA	Cincinatti (OH)	Landfill gas	Gasgrid		PSA	10000	1986
	Dallas (TX)	Landfill gas	Gas grid		PSA	10000	2000
	Dayton (OH)	Landfill gas	Gas grid		Krysol (methanol)	6000	2003
	Houston (TX)	Landfill gas	Gas grid		Selexol	9400	1986
	Los Angeles (CA)	Landfill gas	Vehicle gas		Membrane	2600	1993
	Pittsburg - Monroeville (PA)	Landfill gas	Gas grid		Membrane	5600	2004
	Pittsburg - Valley (PA)	Landfill gas	Gas grid		Membrane	5600	2004
	Renton (WA)	Sewage sludge	Gas grid	98	Water scrubber	4000	1984+1998
	Shasnee (KS)	Landfill gas	Gas grid		Organic physical scrubber	5500	2001
	Staten Island (NY)	Landfill gas	Gas grid		Selexol	13000	1981
United Kingdom	Albury	Landfill gas	Vehicle gas		PSA/Membrane		2008

List of biogas upgrading plant providers

Company	Technology	Website
Acrona-Systems	PSA	www.acrona-systems.com
Air Liquide	Membrane	http://www.airliquide.com
CarboTech	PSA, chemical absorption	http://www.carbotech.de
Cirmac	PSA, Chemical absorption, membrane	www.cirmac.com
Flotech Sweden AB	Water scrubber	www.flotech.com
Gasrec	PSA/Membrane	www.gasrec.co.uk
GtS	Cryogenic	www.gastreatmentservices.com
HAASE	Organic physical scrubbing	www.haase-energietechnik.de
Läckeby Water Group AB	Chemical absorption	www.lackebywater.se
Malmberg Water	Water scrubber	www.malmberg.se
MT-Energie	Chemical absorption	www.mt-energie.com/
Prometheus	Cryogenic	www.prometheus-energy.com
Terracastus Technologies	Membrane	www.terracastus.com
Xebec (QuestAir)	PSA	www.xebecinc.com

An updated version of plant providers can be found on www.iea-biogas.net.

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Abbreviations and definitions

AD	Anaerobic digestion
Biogas	Gas formed during anaerobic digestion by microorganisms of organic material. The gas contains mainly methane and carbon dioxide, but also e.g. hydrogen sulphide. The gas is usually saturated with water.
Biomethane	A gas containing mostly methane produced from a renewable source.
Methane loss	The amount of methane that does not end up in the biomethane stream. Usually expressed as amount lost per total methane gas treated.
Methane slip	Methane that is lost to the atmosphere in the upgrading process. Usually expressed as amount lost per total methane gas treated.
Nm ³	The volume of biogas at 0°C and 1 atm.
Off-gas	The gas that is left when methane has been separated from the biogas in the upgrading process. Contains mostly carbon dioxide, but in general also small amounts of methane.
Raw gas	Untreated biogas.
TWh	Terawatt hours (10 ⁹ kWh)
VS	Volatile solids

