Biomethane production from anaerobic co-digestion at wastewater treatment plants: A critical review on development and innovations in biogas upgrading techniques

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Luong N. Nguyen[a*], Jeevan Kumar[a], Minh T. Vu[a], Johir A. H. Mohammed[a], Nirenkumar Pathak[a], Audrey S. Commault[b], Donna Sutherland[b], Jakub Zdarta[c], Vinay Kumar Tyagi[d], Long D. Nghiem[a,e]

[a] Centre for Technology in Water and Wastewater, School of Civil and Environmental Engineering, University of Technology Sydney, NSW 2220, Australia

[b] Climate Change Cluster (C3), University of Technology Sydney, NSW 2007, Australia

[c] Institute of Chemical Technology and Engineering, Faculty of Chemical Technology, Poznan University of Technology, Berdychowo 4, PL-60965 Poznan, Poland

[d] Environmental Biotechnology Group (EBiTG), Department of Civil Engineering, Indian Institute of Technology Roorkee, 247887, India

[e] NTT Institute of Hi-Technology, Nguyen Tat Thanh University, Ho Chi Minh City, Vietnam

*Corresponding author

Luong N. Nguyen, Centre for Technology in Water and Wastewater, School of Civil and Environmental Engineering, University of Technology Sydney, NSW 2007, Australia (E-mail: luongngoc.nguyen@uts.edu.au)

Abstract

Anaerobic co-digestion (AcoD) can utilise spare digestion capacity at existing wastewater treatment plants (WWTP) to generate surplus biogas beyond the plant's internal energy requirement. Data from industry reports and the peer-reviewed literature show that through AcoD, numerous examples of WWTPs have become net energy producers, necessitating other high-value applications for surplus biogas. A globally emerging trend is to upgrade biogas to biomethane, which can then be used as town gas or transport fuel. Water, organic solvent and chemical scrubbing, pressure swing adsorption, membrane separation, and cryogenic technology are commercially available CO_2 removal technologies for biogas upgrade. Although water scrubbing is currently the most widely applied technology due to low capital and operation cost, significant market growth in membrane separation has been seen over the 2015-2019 period. Further progress in materials engineering and sciences is expected and will further enhance the membrane separation competitiveness for biogas upgrading. Several emerging biotechnologies to i) improve biogas quality from AcoD; ii) accelerate the absorption rate, and iii) captures CO_2 in microalgal culture have also been examined and discussed in this review. Through a combination of AcoD and biogas upgrade, more WWTPs are expected to become net energy producers.

Keywords: Biogas upgrading; Anaerobic co-digestion; Biomethane; Biogas utilisation; Bioenergy.

1. Introduction

Securing affordable and clean energy from sustainable sources is a global challenge of our time. Addressing this challenge has resulted in a paradigm shift in many aspects of the economy, including organic waste management. The conventional view of waste as a disposable material is no longer suitable. In a circular economy, organic waste is a resource for energy and nutrient recovery. Indeed, carbon, nitrogen, phosphorus, and energy can be sustainably and economically extracted from organic wastes such as food wastes, sewage sludge. A globally emerging practice is to valorise urban organic waste via anaerobic co-digestion (AcoD) using the spare capacity at wastewater treatment plants (WWTPs) (Nghiem et al. 2017, Xie et al. 2018, Miryahyaei et al. 2020, Chan et al. 2019, Batlle-Vilanova et al. 2019).

Recent success in full-scale AcoD implementation demonstrates the potential role of WWTPs as energy producers. Anaerobic digestion facilities at WWTPs are used to treat sewage sludge with low organic content. Thus, their capacity is governed by hydraulic rather than organic loading. To utilise the spare digestion capacity (Schwarzenbeck et al. 2008), organic waste can be co-digested with sewage sludge to increase biogas production.

AcoD increases biogas production by 2.5 to 4 times compared to the digestion of only sewage sludge (Shen et al. 2015). Several WWTPs have become net energy producers (Nghiem et al. 2017, Shen et al. 2015, Macintosh et al. 2019). The Grevesmuhlen WWTP (Germany) converts a mixture of primary sludge, waste activated sludge, and grease to biogas, then through gas engines, produces 20% surplus energy (Schwarzenbeck et al. 2008). The Köhlbrandhöft plant (Germany's largest WWTP, serving 1.85 million residents in Hamburg) has also produced 15% more electricity than it has consumes on an annual basis. Encouraging success in AcoD implementation at WWTPs has become an impetus for new applications of the surplus biogas.

Raw biogas contains about 65% CH_4, 35% CO_2, and a trace quantity of hydrogen sulfide, water vapor, ammonia, and siloxane depending on the types of feedstock and digestion process (Mattioli et al. 2017, Wickham et al. 2018, Jang et al. 2015, Martínez et al. 2012). The presence of CO_2 and other trace gases reduces the economic value and limits beneficial applications of biogas. Thus, biogas must be pretreated to remove hydrogen sulfide, water vapor, and other trace gases before the most beneficial applications. In addition to pretreatment, high-value applications such as transport fuel or natural gas grid injection require complete removal of CO_2 for biomethane production. The process of CO_2 removal to produce biomethane is called biogas upgrading. Biogas upgrading technologies such as water or organic physical scrubbing, chemical scrubbings, pressure swing adsorption, membrane separation, and cryogenic technology are available for commercial applications but can be very energy-intensive. The selection of both pretreatment and upgrading technologies depends on the biogas composition, the available resources, and the final product quality.

This paper reviews the state-of-the-art knowledge on the biomethane production processes that can combine with AcoD process at WWTPs to leverage existing infrastructure. This review focuses on the biogas production capacity of AcoD and the potential utilisation of biogas and the associated quality requirements. A major focus is given to the pre-treatment and upgrading technologies since they are essential for beneficial utilisation of the produced biogas. Additional benefits emerging from these techniques are also reviewed. This critical review expects to guide practitioners, water engineers, and scientists on future sustainable development endeavours.

2. Anaerobic co-digestion at WWTPs

2.1 AcoD at WWTPs

AcoD at WWTPs refers to the digestion of sewage sludge with one or more co-substrates with high organic content. These co-substrates are essential organic waste such as food and kitchen waste, organic fraction of municipal solid waste (OFMSW), fat oil and grease (FOG), food/beverage processing waste, and biofuel by-products (i.e., crude glycerol, microalgae, corn silage). The theoretical principle of AcoD is the complementarity between nutrient-rich sewage sludge and carbon-rich organic wastes to boost the anaerobic digestion (AD) performance (Xie et al. 2018, Mattioli et al. 2017, Solé-Bundó et al. 2019, Salama et al. 2019, Siddique et al. 2018, Wang et al. 2020, Aichinger et al. 2015).

AcoD of sewage sludge with organic wastes significantly increases the organic loading rate (OLR) with only a marginal increase in hydraulic loading to enhance biogas production (Wickham et al. 2018, Nghiem et al. 2014a). Nghiem et al. (2014a) demonstrated in a pilot-scale AcoD that intermittent injection of crude glycerol (i.e., byproducts from oil refinery industry) at 0.63 and 3% v/v in sewage sludge led to an increment of 50 and 80% in biogas production. Co-digestion of soft drink beverage waste at 10 and 20% of feed volume increased biogas production by 89 and 191%, respectively (Wickham et al. 2018). Cavinato et al. (2013) assessed both the pilot and full-scale AcoD of sewage sludge and OFMSW and achieved an enhancement in biogas production by nearly 40-50%. Kim et al. (2011a) reported that an 80% increase in the biogas production was attained at WWTP in Velenje, Slovenia. An uplift in specific biogas production was observed at 230%, resulting in a 130% increase in electricity production and 55% in heat energy (Zupančič et al. 2008). Koch et al. (2015) reported that 78% of the energy requirement of WWTPs could be gained from AcoD of sewage sludge with food wastes at ratio of 90:10% feed volume. AcoD at WWTPs have provided 100% required energy in a number of examples (Table 1). Based on the biogas production, AcoD enhances the utilisation of digester volume by 2.5 to 4 times. Shen et al. (2015) reported that AcoD plants produce the biogas at the rate of 2.5 to 4 m^3 per m^3 digester volume compared to 0.9 to 1.1 m^3 in anaerobic digester (AD) plants. Likewise, Wickham et al. (2018) suggested that AD of sewage sludge can receive an additional 2 kg chemical oxygen demand (COD)/m^3.d from beverage waste to achieve an OLR of 3.8 kg COD/m^3.d with proportional increase in biogas production.

Financial benefits from AcoD can be realised through energy production and gate fee. Electricity and heat generated from biogas can be used for onsite consumption. Excess energy can also be sold. About $2 million per year in electricity revenue was achieved through AcoD of fat-oil-and-grease, food waste, and sewage sludge at the East Bay Municipal Utility District, USA (Shen et al. 2015). Another example is to utilise the produced biogas in an adjacent facility to WWTPs, minimising gas transportation and investment cost. The Des Moines Metropolitan Wastewater Reclamation Authority WWTPs (Iowa, USA) sells 40-50% of the produced biogas to a nearby oilseed processing facility, providing an income of 0.8 million USD per year (Zhu et al. 2015).

Gate fee (i.e. or tipping fee – a charge upon a given quantity of waste at waste processing facility) can also generate revenue to support AcoD. In the US, the food waste tipping fee varies from 50 to 170 USD/ton (Shen et al. 2015). In Australia, gate fee typically consists of landfill levy (which is then reinvested to activities that divert waste away from landfill) and operation cost. The current high landfill levy and potential increase in near future (i.e., significant landfill shortages) will create greater incentives for co-digestion of residual municipal solid waste. As an example, the estimated gate fee in New South Wales is $110 USD/ton (Source from Australian Paper's Energy from Waste feasibility study – Fact Sheet 6). Although numerous WWTPs have adapted AcoD in their operation (Table 1), economic data are commercially sensitive and thus rarely available in the literature. Several technical aspects, considerations as well as possible solutions raised from implementation of AcoD at WWTPs have been available in the literature (Nghiem et al. 2017, Xie et al. 2018, Solé-Bundó et al. 2019, Salama et al. 2019, Siddique & Wahid 2018), in the favour of supporting AcoD.

Table 1: Examples of AcoD implementations at WWTP to achieve 100% energy self-sufficiency and become net energy producers (Nghiem et al. 2017, Shen et al. 2015)

Location	**Feedstock (V/V ratio)**	**Capacity (m^3)**	**Biogas production (GWh/y)**
Point Loma WWTP – USA	Mixed PS + WAS	8 x 13,600	193
Gloversville–Johnstown Joint WWTP – USA	Sludge + (yogurt/cheese whey wastewater	1st : 5700 2nd: 4900	28
Sheboygan Regional WWTP – USA	Sludge + FOG + dairy waste	N/A	32
Gresham WWTP – USA	Sludge (87%) + FOG (13%)	2 x 3800	17.2
East Bay Municipal Utility District WWTP - USA	Sludge + FOG/Food waste/HSW	12 x 7500	90
Strass im Zillertal WWTP – Austria	Mixed BNR WAS + Grease trap + Crude glycerol + Food waste	N/A	10
Grevesmuhlen WWTP – Germany	PS (10%) +WAS (60%) + Grease skimming sludge (30%)	2 x 1000	1.95
Zürich Werdhölzli WWTP – Switzerland	Sludge (79%) +FOG (21%) = 23,000 t TS/y	4 x 7250	41.4

WAS = waste activated sludge; PS = primary sludge; FOG = fat oil and grease; BNR = biological nutrient removal; TS = total solid; HSW = high strength waste

2.2. Utilisation of biogas

Most of the produced biogas is currently utilised for heat and electricity generation (Fig. 1). Biogas upgrade to biomethane has only been implemented in a few countries for transport fuel and natural gas grid injection. A notable example is Sweden, where more than half of the produced biogas is used as a transport fuel, supporting 44,000 light vehicles, 750 buses, and 2,200 trucks (data in 2017 from CNG Europe). Germany is currently the world largest biogas producer. Thus, although a small portion of biogas is purified and used as transport fuel, it is enough to power about 96,000 light vehicles, 1,700 buses, and 200 trucks (data in 2017 from CNG Europe). An emerging biomethane market has also been seen in several countries such as Denmark, France, Switzerland, and South Korea (Fig. 1).

Biogas utilisation options are supported initially by government incentives such as feed-in tariffs and tax exemptions, and energy policy. For example, the feed-in tariffs for electricity resulted in biogas being used to produce electricity in Germany, UK, and Austria. Unlike Sweden, the tax exemption favours the transport fuel application. France, Denmark, Sweden, and the UK have strong financial support for biogas injection into gas grids.

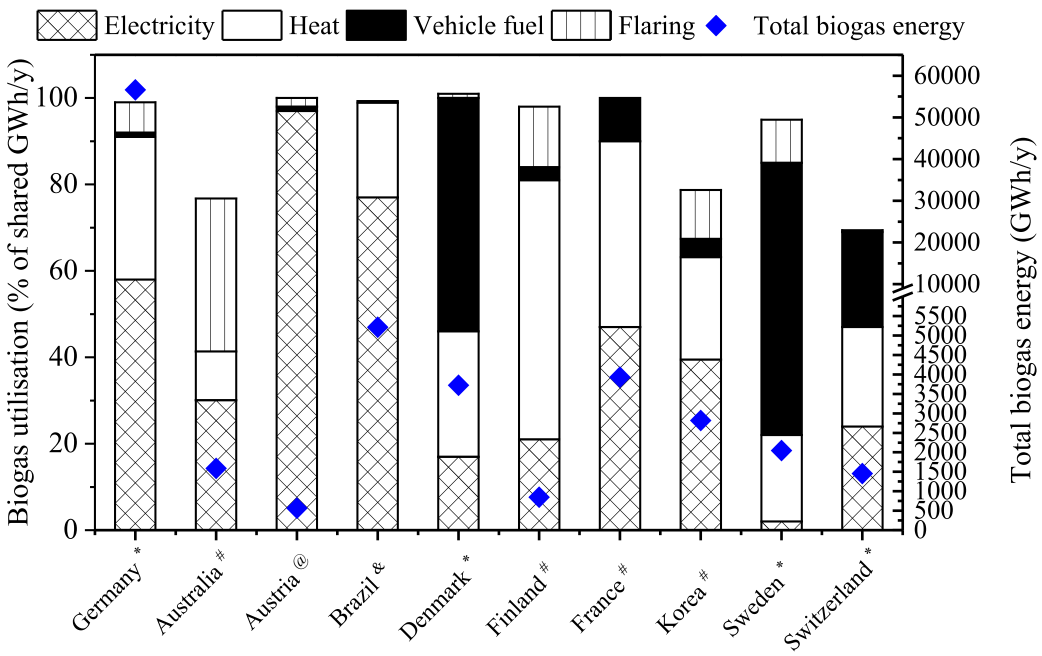

Figure 1: Biogas utilisation (i.e. % of shared GWh/y) in some countries. The symbol *, #, &, @ indicated data source from 2018, 2017, 2016, and 2013, respectively (Source: IEA Bioenergy Task 37).

Wastewater treatment is an energy intensive process. The process accounts for about 3% of consumed electrical energy annually in USA (Wan et al. 2016). It is estimated that the energy demand for wastewater treatment is between 20 and 30 KWh per person annually. The wastewater treatment process also contributes to 5% of global greenhouse gas emission (Nghiem et al. 2017, Gude 2015). In this regard, AD of sewage sludge can produce biogas to compensate 15 to 18 KWh per person. Biogas conversion to heat and energy also reduce the greenhouse gas emission volume at WWTP. Current approach is to intensify the capacity of AD facility via AcoD at WWTP to produce more biogas. Indeed, WWTPs produce a significant volume of biogas (Table 2). For example, WWTPs in Germany contributes above 50% of total biogas production in 2019. It is expected that the amount of biogas production will exceed the heat and energy requirement onsite, necessitating other applications for this renewable energy.

Table 2: Number of WWTPs with AD and AcoD of sewage sludge with organic waste for biogas production and the relative biogas production (Source: IEA Bioenergy Task 37: https://task37.ieabioenergy.com/country-reports.html).

Country	**WWTPs with AD**	**WWTPs with AcoD**	**Biogas production at WWTPs (GWh/y)**	**Biogas from WWTPs (% of total production)**	**Year of data collection**
Australia	52	2	381	24	2017
Brazil	10	3	210	4	2016
Denmark	51	n.a	308	8.3	2018
Finland	16	n.a	162	23	2017
France	88	n.a	442	26	2017
Germany	1274	n.a	3657	56.2	2019
Norway	27	n.a	240	33	2015
South Korea	78	21	630	22	2017
Sweden	138	n.a	715	35	2018
Switzerland	473	n.a	633	43.5	2018
The Netherlands	80	n.a	640	37.2	2018
United Kingdom	163	n.a	1280	15.4	2018
Canada	31	n.a	n.a	20.7	2019
USA	1241	216	n.a	n.a	2015

Providing electricity to the power grid or injecting biomethane to the natural gas grid for distribution and transport fuel are potential applications of the surplus biogas at WWTPs. Feeding electricity to the power grid is not always feasible. There have been some government incentives especially Europe that allows WWTPs to feed surplus electricity to the power grid at a favourable tariff. However, many of these incentives have expired or about to expire. In some countries, WWTP utilities may not have a power generator license to inject electricity into the power grid. There are also major technical challenges to the existing energy infrastructure, which was originally designed only for energy distribution rather than a flexible feed in and sharing network. Synchronisation of multiple power sources to the distribution grid needs to match the voltage, frequency, and phases. In addition, the imbalance between supply and demand as well as inappropriate load management can destabilise the distribution grid.

Instead of electricity production, upgrading to biomethane for domestic consumption and transport fuel appears to be an appealing alternative for the surplus biogas from WWTPs. The methane economy is mature and many countries have extensive natural gas grid distribution. The surplus biomethane production at WWTPs can be fed into the gas grid for distribution and storage. This also takes the advantages that storage capacity and duration of storage for methane is significantly higher than other energy storage systems (e.g. electricity in battery). Facilities such as natural gas refuel stations and increase in number of natural gas vehicles in line with the production of biomethane. An example of a complete production line includes biogas production, biogas upgrading and a refuelling station has been operated at a Swedish WWTP. Thus, biogas upgrading to biomethane for natural gas grid injection or transport fuel is probably a preferred option for the surplus gas at WWTPs.

3. Methane and other gases in biogas

Raw biogas typically contains about 65% CH_4 and 35% CO_2 (Mattioli et al. 2017, Wickham et al. 2018). The energy content of biogas is defined by the methane concentration – the higher the methane, the higher the calorific energy value of the gas. For example, the calorific value (i.e., Wobbe index) of biogas with 70% of CH_4 content is 21.5 MJ/Nm^3, whereas that of biomethane (100% CH_4) is 35.8 MJ/Nm^3. The high volume of CO_2 in biogas does not only reduce the calorific but also make it uneconomic for compression and transportation for off-site utilisation.

Trace gases especially hydrogen sulfide (H_2S), water vapor, and siloxane can be detrimental to downstream biogas utilisation processes. H_2S is corrosive to co-generators, biogas storage facilities, compressors, and pipelines. The combustion of H_2S produces sulfur dioxide, which is a major air pollutant (Zhu et al. 2015).

H_2S level in biogas from AcoD is an important parameter affecting the usage of biogas. Under anaerobic conditions, sulphur-bearing organic compounds and sulphate in organic wastes are reduced to sulphide, which is released to biogas in the form of H_2S (Cirne et al. 2008) . High sulphur content substrate produces high H_2S content in biogas (Hansen et al. 2004) . The H_2S content in biogas from a WWTP digesting only sewage sludge is in the range from 500 to 2,500 ppmv. Animal wastes, slaughterhouse wastes, dairy milk industry wastes can produce biogas with 20,000 to 30,000 ppmv H_2S (Hansen et al. 2004). Up to 31,000 ppmv H_2S in biogas from AcoD with seaweed has been reported (Hansen et al. 2004). By contrast, most internal combustion engines manufactures limit H_2S to 100 ppmv in biogas. According to the European biomethane standards, the concentration of H_2S is required to be less than 1 ppmv for gas grid injection and transport fuel (European biomethane standards).

Water vapor in biogas can also lead to corrosion problems (Ryckebosch et al. 2011) . Water content in biogas depends on the digester's operating temperature (e.g., mesophilic or thermophilic). The water content is about 4 to 5% (v/v) of raw biogas from the mesophilic digester. At higher temperatures (i.e., 55 to 60°C in thermophilic digester), 7 - 8% (v/v) (between 30 and 100 g water per m^3) of the water content has been recorded. Water vapor removal is necessary to avoid corrosion in biogas upgrading and utilisation. The permissible water content is below 10 mg/Nm^3 for gas grid injection.

The presence of siloxanes in biogas can lead to the formation of siloxane dioxide particles. Siloxane dioxide particles are abbrasive and are adhessive to metal surface, causing excessive wear and tear of the the co-generator engines. Siloxanes concentration in biogas is between 1 to 400 mg(Si)/Nm^3, while the maximum permissibl siloxane concentration in natural gas is 5 mg (Si)/Nm^3 (Ryckebosch et al. 2011).

Biomethane for gas grid injection and transport fuel must meet very stringgent standards and are due to the relevance of the technical execution of installations, planning, construction, and operation. The European Commission has begun to develop the European biomethane standards for grid injection and transport fuel. The new standard is set to bring legal and technical security, market assessment, and precondition free trade amongst countries. Requirements such as total H_2S < 1 ppmv, and siloxanes < 0.5 mg(Si)/Nm^3 are examples in the new standards.

4. Biomethane market

Biomethane market has gained significant momentum in recent years. The number of new biogas upgrading plants increases worldwide (Table 3). It results from combined factors including i) advanced in biogas upgrading technology; ii) paradigm shift from a low economic electricity and heat production to new opportunities for use biomethane in the transport sector, and iii) moving towards a green economy model (Zhu et al. 2019). As an upgraded product of biogas, biomethane is essentially identical to natural gas. Thus, biomethane can be injected into the natural gas grid or used as transport fuel. Germany and Sweden have the largest markets for biomethane in the world. A growing interest can also be seen in other countries, especially the UK, France, and Switzerland (Table 3).

The global biomethane market was valued at USD 0.62 billion in 2017. With the annual growth rate of 26%, a $4.96 billion market size is estimated by 2026. Several countries have set ambitious target for biomethane as natural gas replacement for household consumption (Hoo et al. 2020). France plans to provide 8 TWh of biomethane energy by 2023 (Herbes et al. 2018). In the UK, biomethane is expected to be a major source of the future clean gas supply (Richards et al. 2020).

Biomethane can also be liquefied or compressed biomethane for storage or used as transport fuels. Liquefied biomethane also has higher energy content, suitable for heavy vehicles and providing long distance transportation. Biomethane utilisation as transport fuel has continued to increase over the years. Sweden has set a target of 100% of transport fuel from biomethane by 2030. This target appears realistic. In 2016, 82.8% of transport fuel was from biomethane. This number increased to 90.8% in 2018.

Table 3: Increase in the number of biogas upgrading plants in selected countries over the 2014 – 2019 period (Source: IEA Bioenergy Task 37: https://task37.ieabioenergy.com/country-reports.html).

Country	Number of plants in 2014	Number of plants 2016	Number of plants in 2019
France	8	30	47
Denmark	12	32	34
United Kingdom	37	85	96
Italy	5	7	8
Finland	9	12	17
Switzerland	24	31	45
Netherlands	21	26	53
Germany	178	194	203
Austria	14	15	13
Sweden	59	63	69
Hungary	2	2	n/a
Luxembourg	3	3	3
Spain	1	1	
Norway	n/a	n/a	9
Australia	0	0	0
South Korea	n/a	n/a	10
Japan	n/a	6	6
China	n/a	n/a	2
USA	n/a	n/a	50

n/a = not available

5. Biogas pretreatment

5.1 H_2S removal

Desulphurisation of raw biogas is considered the essential step before further processing and the use of biomethane. Mothods to remove H_2S from biogas can be categorised into three groups: i) biological desulphurisation; ii) absorption to a liquid solution (water or chemical scrubbing), and iii) adsorption on a solid absorbent (iron sponge, iron oxide pellets, activated carbon).

Biological desulphurisation can be performed *in-situ* to the anaerobic digester. Air or oxygen is injected into the digester to provide oxygen molecules (Ryckebosch et al. 2011, Nghiem et al. 2014b). Nghiem et al. (2014b) injected oxygen to regulate the oxidation redox potential between -320 to -270 mV to reduced H_2S in biogas from 6000 to below 30 ppm without any observable effect on digester performance. Biological desulphurisation can be carried out ex-situ in biofilters, which are packed bed scrubbers containing immobilized microorganisms. H_2S is captured in the liquid film and biologically translated to sulphur and sulphate. The liquid of the scrubber bed can be regenerated if the pH level decreases below 7. This system is unable to supply a stable H_2S content <100 ppmv, and this value varies with the qualities of the raw biogas. Hence, this process can not be confidently employed for biomethane production (Petersson et al. 2009).

H_2S in biogas can be removed by absorption in the scurbbing technologies (i.e., physical and chemical scrubbing) (Table 4). Water and alkaline solution (e.g., sodium hydroxide, calcium hydroxide, and potassium hydroxide) and amines can absorb H_2S. In this regard, H_2S can be simultaneously removed during biogas upgrading (i.e. CO_2 removal). However, it is worth mentioning that chemical reaction between H_2S with absorbent is an irreversible process, limiting the absorbent regeneration.

Adsorption of H_2S in iron oxide pellets/sponge, and activated carbon column is an effective method for biogas treatment. Iron oxide reacts with H_2S in biogas to form ferric sulfide (Wang et al. 2011). Wang et al. (2011) reported that iron oxide could uptake large amount of H_2S (e.g., 0.25 kg H_2S per kg iron oxide). The formed ferric sulfide can be changed to ferric oxide and elemental sulfure by exposing it to air or oxygen during the regeneration process. Adsorption is the most applied method for H_2S removal because of its outstanding performance, regeneration capacity, and easy to use.

Table 4. Considerations for selection of desulphurisation techniques

Method	Considerations	References
Air/oxygen injection	- Potential over oxygenation affect anaerobic conditions - High cost of pure oxygen bottles - Limited full scale experience - High volume of N_2 in biogas	(Nghiem et al. 2014b, Jeníček et al. 2017)
Bio-trickling filter	- Unstable performance - Slow reaction rate - Difficult to set up the filter	(Montebello et al. 2013)
Water absorption (scrubbing)	- High water volume - Unstable performance - Can remove part of CO_2	(Angelidaki et al. 2018)
Chemical absorption (scrubbing)	- Ongoing chemical cost (no regeneration) - Performance is predictable - Partial CO_2 removal	(Ryckebosch et al. 2011)
	- Addition of catalyst solution (Fe(III)-EDTA to reduce chemical consumption	(Horikawa et al. 2004)
Iron sponge (Iron oxide/hydroxide) adsorption	- High operating costs - Excessive heat generation - H_2SO_4 formation	(Angelidaki et al. 2018)
Activated carbon adsorption	- H_2SO_4 formation - Impregnated with NaOH, KOH, Na_2CO_3 - Regeneration requirements more frequent - Modification of AC with $CuSO_4$, KOH, $ZnAc_2$	(Zulkefli et al. 2019, Ciahotný et al. 2019)

5.2 Water vapour removal

There are several methods to remove water vapour. Biogas can be cooled down to allow water vapor to condense (Ryckebosch et al. 2011). The condensed water is returned to the digester or drained out. Moisture in biogas can also be removed by adsorption dryers. These are high water adsorbent materials (e.g. silica gel, aluminum oxides, and molecular sieves). The used adsorbent can be regenerated by heating. Moisture can also be removed by increasing the biogas pressure. In this process, water vapor is not completely removed but the relative humidity of biogas is reduced. It is also noted that water vapour removal is performed after CO_2 removal in scurbbing technologies.

5.3 Ammonia removal

Ammonia can be present in biogas at a trace level of up to 100 ppm (Ryckebosch et al. 2011). Ammonia can be removed simultaneously with water because of its high solubility in water. Scrubbing technologies (i.e. physical and chemical scrubbings) are effective in achieving complete removal of ammonia from raw biogas (Allegue et al. 2012). Therefore, the pretreatment to removal of water vapor and ammonia is not required if these methods are applied during CO_2 removal (i.e. biogas upgrading).

5.4 Siloxanes removal

Absorption and adsorption are the two common methods for the removal of siloxanes from biogas. Organic solvents, strong acids or bases solution can provide upto 97% siloxanes removal efficiency (Ryckebosch et al. 2011). However, the use of organic solvents, acids or bases could cause corrosion and produce hazardous chemicals that requires additional treatment. Adsorption on solid materials such as silicagel and activated carbon is preferred option. Activated carbon adsorption reduces siloxanes level to 0.1 mg/Nm^3 (Ajhar et al. 2010). Adsorbent regeneration is prossible at high temperature (i.e., above 250 °C). Siloxanes removal is usually performed after water vapor since high moisture gas can decrease the removal efficiency (Schweigkofler et al. 2001).

6. Biogas upgrading technologies

Once pre-treated, biogas can be further processed to remove CO_2 to produce biomethane. Several biogas upgrading methods are already available at commercial scale. They include scrubbing (i.e. water, organic solvent, and chemical scrubbing), pressure swing adsorption, membrane separation, and cryogenic technology. The maturity of these methods varies widely. It is noteworthy that some biogas upgrade methods can result in the removal of impurities, especially H_2S. For example, water scrubbing (at high pH) and pressure swing adsorption can remove both H_2S and CO_2 together. On the other hand, a pretreatment step to remove H_2S is required for CO_2 removal by chemical scrubbing using amines. H_2S removal is also required to avoid membrane poisoning in membrane separation techniques.

The high CH_4 purity is required for natural gas gird injection and vehicle fuel, meeting a few criteria such as high-energy content, gas transportation, storage, and technical restrictions. For example, biomethane is compressed in pressurised gas cylinders at 200 to 250 bar for storing and transporting purposes. While CH_4 can be readily compressed, a mixture of CO_2 and CH_4 has very different thermodynamic properties and cannot be readily compressed at high pressure for storage.

6.1 Scrubbing Technologies

6.1.1 Water or organic physical scrubbing

Water or solvent scrubbing relies on the difference in the solubility of gasses (CO_2 and CH_4) in a wash solution (Andriani et al. 2014). The wash solution can be water (water scrubbing) or organic solvent (e.g. polyethylene glycol dimethyl ether, trade name as Genosorb or Seloxol). This method involves no chemical reaction. Since the gas solubility improves with increasing pressure, pre-treated biogas is pressurised and injected into the scrubbing column (Fig. 2).

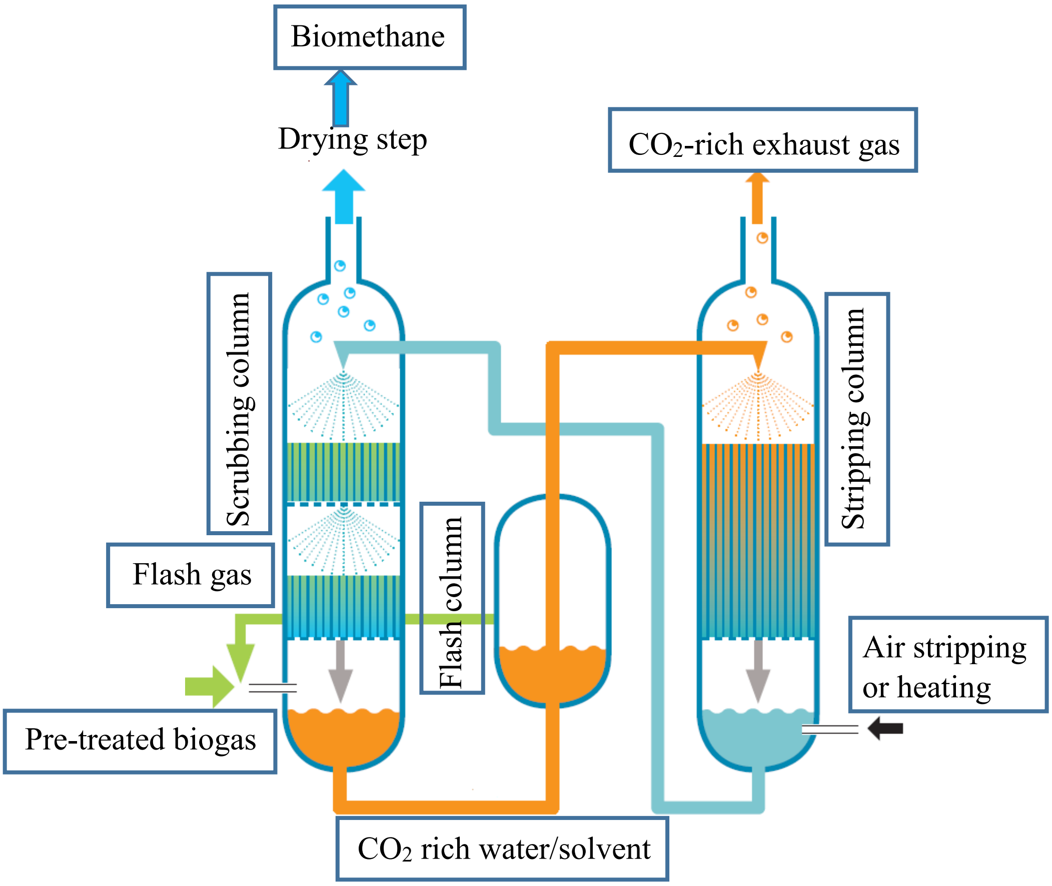

Figure 2: Schematic of scrubbing technologies for the separation of CO_2 and CH_4

In the water scrubbing process, the pretreated biogas is maintained 6-10 bar and 40 °C. At this condition, the solubility of CO_2 is approximately 26 times higher than that of CH_4. The gas is injected via the bottom side of the column, while water is provided from the top. The counter-current injection increases the gas and water interaction in the scrubbing column. This configuration also allows CH_4 venting out from the top while CO_2 rich water is circulated into a flash column from the bottom. At the flash column, the gas pressure decreases to 2.5 – 3.5 bars, releasing CH_4 gas to be recovered. The CO_2 rich water is pumped into a stripping column. In this column, the air is injected at atmospheric pressure, resulting in the removal of CO_2 from water (i.e., regeneration). The ventilated CH_4 is subjected to a drying step to produce final biomethane (Angelidaki et al. 2018).

Although water scrubbing is a simple process with low energy consumption, high water consumption, and methane loss are major drawbacks. A total of 3-5% of methane can be lost, and the combustion of the exhaust gas is required to maintain emission regulation (Ryckebosch et al. 2011) . The water scrubbing method requires a large amount of water (200 m^3/h for a gas flow of 1000 Nm^3/h) (Sun et al. 2015). Thus, water regeneration is crucial for the economic viability of this technology. Water scrubbing can be advantageous when apply at WWTPs. Secondary and tertiary effluent can be used as water source without regeneration (Angelidaki et al. 2018).

An organic solvent can also be used as the wash solution. The process configuration is similar to water scrubbing (Fig. 2). CO_2 has a higher solubility in some solvent such as polyethylene glycol dimethyl ether than in water. Consequently, a smaller volume of solvent is required and the size of the scrubbing column can be reduced. The absorption process also occurs at lower pressure (4 to 8 bars) resulting in a lower energy demand compared to water scrubbing (6 to 10 bars. However, organic solvent regeneration is a complex process compared to water regeneration. Air stripping and pressure release are not effective to regenerate the organic solvents. In practice, organic solvent is heated to between 40 and 80 °C, requiring additional energy of 0.1 to 0.15 kWh/Nm^3 of biogas for regeneration (Ryckebosch et al. 2011, Gupta 2003, Singhal et al. 2017).

Table 5: Advantages and disadvantages of physical scrubbing technologies (Singhal et al. 2017, Kadam et al. 2017, Niesner et al. 2013)

Method	Advantages	Disadvantages
Water scrubbing	- Many years of experience - Numerous plants are under operation - Less costly (i.e. water is a low-cost solvent). - Environmentally friendly solvent - Technically simple method - No additional heat	- Energy consumption: 0.2 to 0.5 kWh/Nm^3 of biogas. - High pressure 4-10 bars - Methane is up to 5 % by volume - Water is less selective (i.e. absorbent rate and loading is low)
Solvent scrubbing	- High absorption rate and loading per volume of solvent - Smaller footprint	- Energy consumption: 0.1 to 0.33 kWh/Nm^3 of biogas. - Additional heat to achieve effective regeneration - Potential environment pollution of used solvent - Methane loss is up to 4 % by volume

6.1.2 Chemical scrubbing

Chemical scrubbing or chemical absorption is based on a reversible reaction between CO_2 with a chemical adsorbent. Common chemical absorbents are monoethanolamine (MEA), diethanolamine (DEA), methyldiethanolamine (MDEA), and other amine compounds. Their solutions have high selectivity against CO_2. Since chemical adsorbents are only reactive with CO_2, the CH_4 loss is minimal after its dissolution in solvent solution (0.1 to 0.2%) (Sun et al. 2015). Thus, a post-combustion of lean gas is not required. Chemical scrubbing can produce high CH_4 purity (99% by volume). H_2S removal upstream must be conducted because of the corrosive reaction of H_2S with amine solution, i.e., degradation of amine (Vega et al. 2014).

Regeneration of an amine solution is an energy-intensive process compared to the physical scrubbing due to the strong binding between the gas molecules. The CO_2 saturated amine solution is heated to above 110 °C for regeneration. The regenerated amine solution then cooled down to 40 °C before starting a new absorption cycle. Regeneration consumes 0.4 to 0.8 kWh/Nm^3 of biogas, or about 15 to 30% of the energy generated from the biomethane (Leung et al. 2014). Recent research has focused on minimising thermal energy requirement for regenerating amine solution. It is achieved by developing a new amine solution, optimising heat-exchanging equipment, and modifying operation conditions (temperature and gas flow rate) (Aroonwilas et al. 2009, Kim et al. 2011b). Amine degradation, equipment corrosion, and potential generation of the volatile compound into the atmosphere are other limitations of the chemical scrubbing method. Moreover, amine can degrade into nitrosamines and nitramines, potentially harmful to human health and the environment (Stowe et al. 2017).

6.2 Pressure swing adsorption

Pressure swing adsorption relies on the principle that CH_4 and CO_2 adsorb differently to specific surfaces or pores of adsorbents. Since the adsorption of CO_2 is proportionally to high pressures and low temperatures, the pressure swing adsorption process utilise pressure/temperature differences, i.e., pressure-temperature swing, to carry out the separation (Ntiamoah et al. 2016). The system main part is a column, filling with adsorbents such as activated carbon, zeolites, calcium oxides, hydrotalcites, and carbon molecular sieves (Fig. 3). These materials are porous and of high surface areas to enhance adsorption capacity (Leung et al. 2014). Desulphurisation is required before adsorption since H_2S is irreversibly adsorbed by the adsorption substance and produces toxic effects (Patterson et al. 2011).

In the pressure swing adsorption, pre-treated biogas is compressed to 2-7 bar and cooled to about 70 °C to improve the adsorption. The pressured gas is injected into the adsorption column from the bottom. CO_2 molecules, which are smaller than methane molecules, accumulate to a much greater degree on the surfaces or in the pores than CH_4. At the same time, CH_4 remains primarily in the gas phase and escapes from the column head, resulting in a methane-rich product gas. Once the methane is released, the pressure inside the column decreases to atmospheric pressure. The adsorbed CO_2 dissolves from the surfaces and returns into the gas phase. This gas is blown off (CO_2- rich exhaust gas) via a valve at the column bottom (Fig. 3).

The column is then filled with biogas to begin a new cycle. Pressure swing adsorption has been in operation for many years at many reference plants for biogas upgrading. The biomethane quality is nearing 96-98%, with 1.5 to 2.5% methane loss (Allegue et al. 2012). Therefore, post-combustion of exhaust gas is required to minimise methane release in the atmosphere (Sun et al. 2015). Overall, the energy requirement of the pressure swing adsorption is between 0.15 to 0.35 kWh/Nm^3 of biogas, making it a competitive method for biogas upgrading.

In the temperature swing adsorption, pre-treated biogas is injected into the column at ambient temperature and pressure allowing CO_2 molecules to adsorb on the materials. Regeneration, on the other hand, is conducted by directly heating the column or injecting hot air, N_2 gas, or steam into the column (Ntiamoah et al. 2016). The regeneration rate is dependent on temperature. Indeed, higher temperature results in a faster regeneration rate. In comparison to pressure swing adsorption, the regeneration time usually is longer. After regeneration, the column is cooled down to ambient temperature. N_2 gas is applied to both cool and clean the column for the next adsorption cycle. Temperature swing adsorption is mainly applied to capture CO_2 from the power station and utilise the wasted heat in the regeneration process.

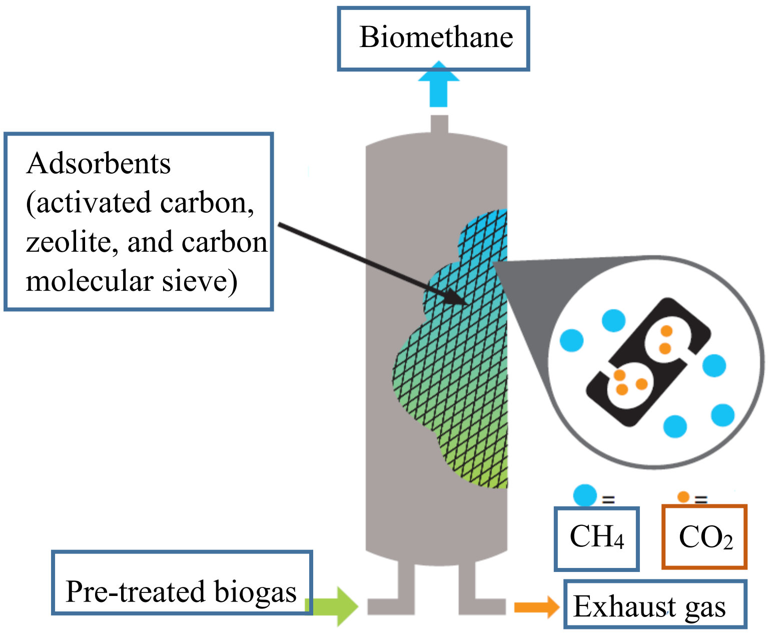

Figure 3. Basic principle of pressure swing adsorption

6.3. Membrane separation

The principle of membrane separation methods is that gases permeate through the membrane pores at different selectivity, i.e., highly permeable to CO_2 (small molecule) and impermeable to CH_4 (large molecule). In general, membrane suitable for biogas upgrading is 20 more permeable to CO_2 than to CH_4. The CO_2-rich exhaust gas from the membrane separation can be used to produce highly pure CO_2 suitable for the food and beverage industry (Esposito et al. 2019). Esposito et al. (2019) evaluated the first large scale industrial biogas upgrading plant to produce CH_4 and CO_2 from membrane separation simultaneously, liquefying and cryogenic units. The residual CO2 from five membranes line was combined and subjected to a liquefying, compression, drying, and cooling. High purity CO_2 (99.9%) was achieved after cooling to -30°C to separate N_2, O_2, and trace CH_4.

Membrane separation is available in different designs. Typical operating pressures are 7 to 20 bars (Peppers et al. 2019). To achieve high methane purities, the tube bundles are connected in two-stage or three-stage cascades. The two-stage cascade provides higher CH_4 and maintains higher recovery than a single cascade. In the two-stage cascade, a circulation loop returns the gas from the first membrane back to the inlet, while the enriched CH_4 continue to the second membrane (Fig. 4).

Key advantages of membrane separation include modular and compact design with less moving parts. However, membrane separation is still an emerging technology with limited practical experience. Moreover, the energy requirement is between 0.18 to 0.33 kWh/Nm^3 of biogas (Makaruk et al. 2010). Methane loss of up to 2% has been reported in some laboratory-scale studies (Baena-Moreno et al. 2020). Peppers et al. (2019) recently investigated the feasibility of membrane separation for 100 Nm^3/h biogas plant. The results demonstrated that pre-treatment of other gas is necessary to protect the membrane and ensure high CH_4 purity (Baena-Moreno et al. 2020). Although biogas quality satisfied the standard, the overall cost analysis revealed low economic viability at small scale (< 100 Nm^3/h biogas flowrate) (Peppers et al. 2019).

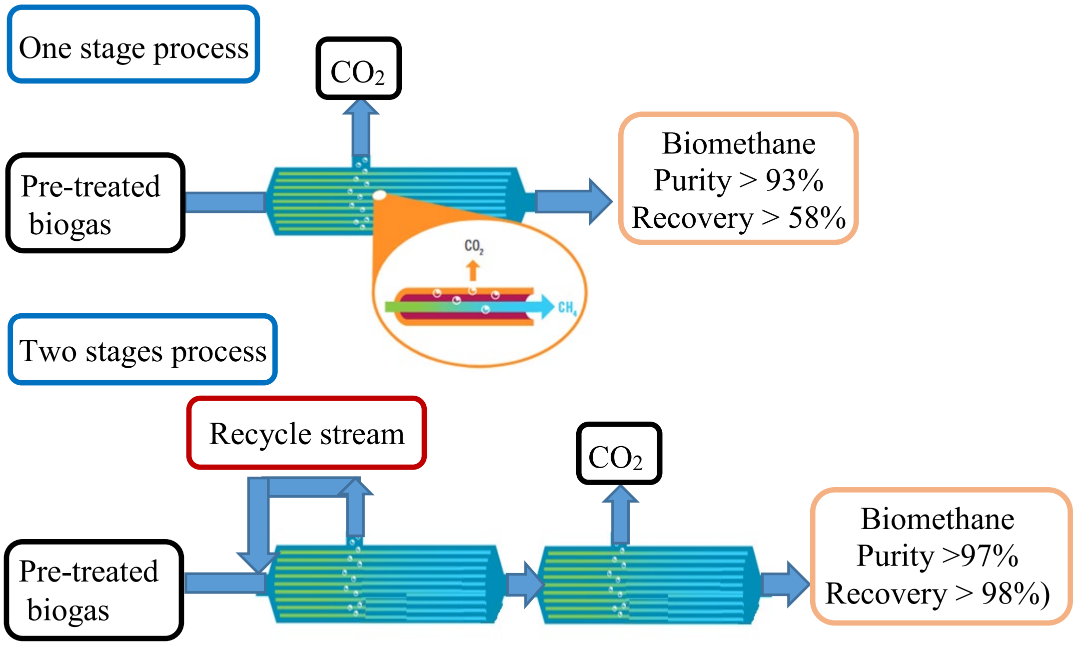

Figure 4: Physical and technical principle of membrane separation

6.4 Cryogenic technology

Cryogenic treatment is based on the principle that gases condense (become liquid) or re-sublimate (become solid) at low temperatures or high pressures. CO_2 and CH_4 have different condensation temperatures. The CO_2 re-sublimates at -78.5 °C and 1 bar while CH_4 remains gaseous. The solid CO_2 and gaseous CH_4 can be separated through rectification (i.e., counter-current distillation). Because of this principle, cryogenic treatment can achieve very pure CH_4 (up to 99.9% by volume), CO_2 (up to 99.9% by volume) with less than 1% methane loss. However, the technology is still under development, and its market readiness has not yet been fully established.

A ubiquitous and significant obstacle to this technology is the high energy required for refrigeration and compression of the raw biogas. The energy consumption is approximately 10% of the generated methane. Another challenge is to ensure that frozen CO_2 does not clog the equipment in the gas refrigeration process. In this regard, other biogas impurities must be carefully removed. However, possible options to strengthen this technology are available. The energy used to condense initial biogas can be recovered if the produced biomethane is to be liquefied. Biomethane liquefaction at -125 °C and 15 bar leverage synergies in the production of cold gas, thus minimising the energy consumption in both steps. Likewise, frozen CO_2 can be utilised as dry ice in some industrial applications (Fig. 5) (Esposito et al. 2019). Thus, cryogenic treatment is starting to become commercially competitive.

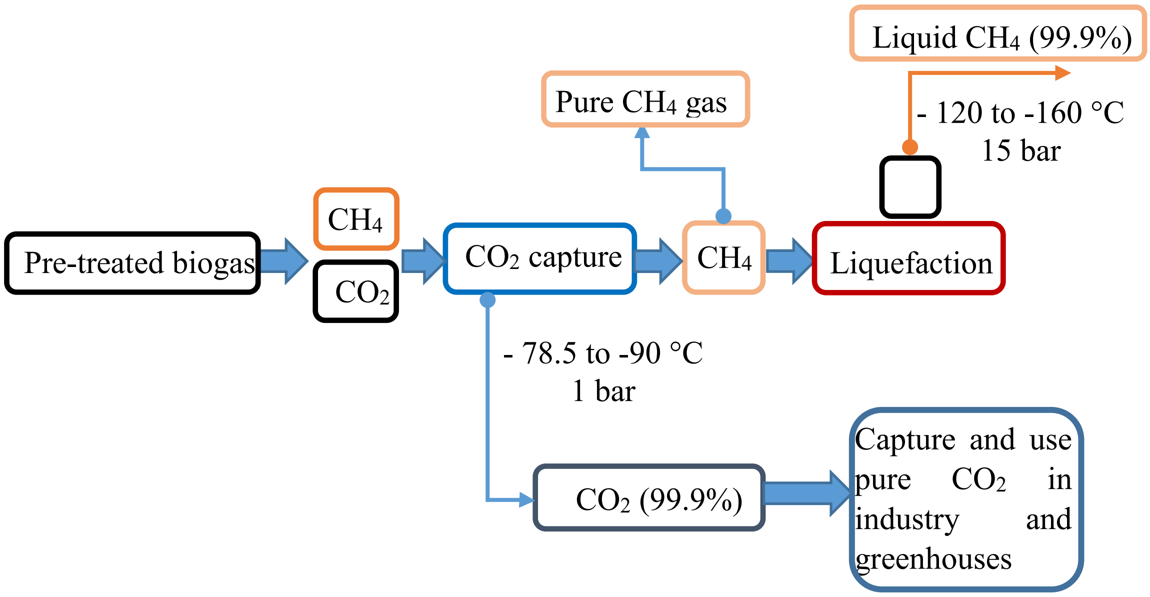

Figure 5. Principle of cryogenic biogas upgrading with potential to capture pure CO_2 and liquefy CH_4

6.5 Current full-scale application

The number of full-scale plants utilising biogas upgrading technologies is increasing (Fig. 6). Physical scrubbing using water (i.e. water scrubbing) is the dominant technology. In 2019, there have been 181 plants in operation. Water scrubbing is a simple process in comparison to others technologies. Its major drawback is high water volume requirement. Reusing secondary or tertiary effluent for scrubbing can reduce overall cost. The market share of membrane separation technology has grown significantly over the last five years. The number of plants increased from 92 (2015) to 173 (2019) (Fig. 6). Key advantages of membrane separation include robust design with less moving parts, modular design, and a small physical footprint. Regent scientific progress in materials engineering and science has also resulted in better membrane performance. With the accumulation of practical experience, it is expected that membrane separation will be highly adopted in the near future. Chemical scrubbing, organic physical scrubbing, pressure swing adsorption, and cryogenic technology have a small number of new instalment over last five years. Cryogenic technology could hold promise for future development once the benefit of pure CO_2 harvesting for dry ice production is realised (Esposito et al. 2019).

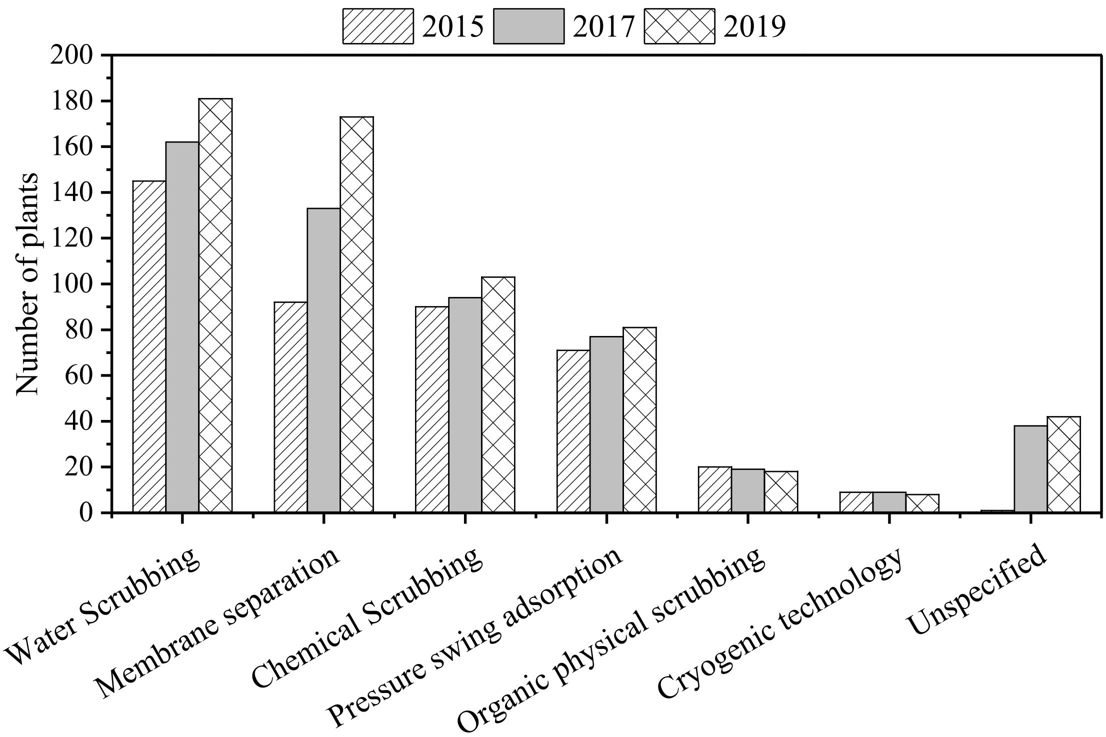

Figure 6: Biogas upgrading plants in countries employing different technologies from over the last 5 years.

The comparison of common biogas upgrading technologies is summarised in Table 6. A direct comparison among these technologies is not possible since their selection can depend on multiple factors beyond those summarised in Table 6. Nevertheless, some generalisation can be made. An estimated OPEX for plant with capacity of 1000 Nm^3/h indicates that water scrubbing and membrane separation have low operating and maintenance costs (Table 6). Membrane separation can also achieve the highest biomethane quality with moderate energy consumption and methane loss. Data in Table 6 are consistent the number of full-scale biogas upgrading plants currently in operation. Water scrubbing and membrane separation are the two most prevalent biogas upgrading technologies (Figure 6). Figure 6 also shows a significant increase in the number of membrane-based biogas upgrading plants over the 2014-2019 period.

Table 6: Reported energy consumption (kWh/Nm^3) of different technologies. Source: (Singhal et al. 2017, Patterson et al. 2011, Masebinu et al. 2014, Vrbová et al. 2017)

Technologies	Biomethane quality (CH_4 %)	Energy consumption (kWh/Nm^3)	Methane loss (vol %)	Cost for 1000 Nm^3/h plant	
				CAPEX (million €)	OPEX (€/year)
Water scrubbing	95–98	0.2–0.5	0.5 - 5	1	15,000
Organic physical scrubbing	93–98	0.1–0.33	1 - 4	1	39,000
Chemical scrubbing	<98	0.05–0.18	0.5	2	59,000
Pressure swing adsorption	<98	0.16–0.43	1.5 - 2.5	1.75	56,000
Membrane separation	90–99	0.18–0.35	0.5 - 2	2	25,000
Cryogenic	99	0.18–0.25	0.1	n.a	n.a

6.6. Emerging biotechnology platforms for biogas upgrading

6.6.1 Technologies to improve biogas quality from AcoD

Biological biogas upgrading targets different microbial functional groups in the AcoD process to facilitate its function to serve a specific aim. Anaerobic digestion is a biochemical process that involves four groups of microorganisms, namely hydrolysers, acidogens, acetogens, and methanogens (Nguyen et al. 2019). Biological desulphurisation is one example that facilitates the function of sulphur-oxidising microorganisms to reduce H_2S in biogas. The success of the biological desulphurisation (Section 3) sets a foundation for further exploration to develop biological biogas upgrading technology. In this regard, the presence of hydrogenotrophic methanogens in the AcoD is of particular interest. Hydrogenotrophic methanogens mainly use H_2 as electron-donating sources for the reduction of CO_2 to methane. Thus, it is hypothesised that through the exogenous addition of H_2 into the digester, CO_2 can be converted to CH_4 to achieve a two-fold benefit: high CH_4 and low CO_2 content in biogas. Wahid et al. (2019) observed that the addition of H_2 at a ratio of 4 to 1 mole of CO_2 resulted in 94 and 3% of CH_4 and CO_2, respectively, in the biogas. Likewise, Bassani et al. (2015) achieved 89 and 85% CH_4 content in biogas from mesophilic and thermophilic digesters, respectively, after H_2 addition.

Although the methane content in the final biogas is higher after H_2 addition, this technology is still at its infancy with results from laboratory-scale studies only. There are many drawbacks. Residual H_2 in the biogas is one limitation. Wahid et al. (2019) observed up to 3% of H_2 in the biogas, which is higher than the biomethane quality standard for natural gas injection and transport fuel. Injection of H_2 into the digester can increase the pH (i.e., due to CO_2 depletion) and influence the process stability (Luo et al. 2013). pH over 8.5 can inhibit the methanogenic activity (Nguyen et al. 2019). In addition, H_2 injection can increase the hydrogen partial pressure that may inhibit the acetogenesis.

6.6.2 Biocatalytic enzyme enhance CO_2 capture efficiency

Research efforts to enhance the efficiency of adsorbents (adsorbent rate and capacity) are in the trajectory to reduce the energy cost of the biogas upgrading technologies. Using enzymes such as the carbonic anhydrases to convert CO_2 and water to bicarbonate (reaction described below), could contribute to reducing the energy cost of the CO_2 removal step. In the chemical absorption method, the energy requirement is determined by the solvent specific heat of reaction and solvent capacity to take up CO_2. Consequently, if a solvent has a low reaction heat and high capacity, energy saving can be achieved (Closmann et al. 2009, Gundersen et al. 2014, Kunze et al. 2015). Amines and alkali carbonates are potential solvent candidates, but they suffer slow absorption kinetics (Kunze et al. 2015, Beiron et al. 2019). Bicarbonate formation is the rate-limiting step of the absorption of CO_2.

Enzymes can act as an activator to enhance the absorption kinetics. Indeed, Kunze et al. (2015) demonstrated that the addition of carbonic anhydrase at 0.2 (wt %) to 30 wt % MEA and K_2CO_3 improved the absorbed volume by a factor > 4. Likewise, MDEA absorption capacity was increased by a factor of 3 after the addition of the carbonic anhydrase enzyme (Vinoba et al. 2013). The pilot-scale testing performed at 70 °C, revealed that enzyme addition was a technically feasible method. Thus, the biotechnology enzyme will help to advance the enzyme addition technology through enhancing temperature resilience. However, to the best of our knowledge, no study has investigated the application of this technology for biomethane production from biogas.

6.6.3 Microalgae for CO_2 capture from biogas

Microalgae are autotrophic microorganisms that can fix CO_2 and utilise nutrients (nitrogen and phosphorus) to produce biomass using light. Microalgae biomass can be used in an array of valuable bioproducts such as food products, nutraceuticals, feed, pharmaceuticals, biopolymers, bioplastics, and bulk chemicals (Fabris et al. 2020, Vu et al. 2020a). Therefore, the application of microalgae to capture CO_2 from biogas can have multi-fold benefits, including i) reduction in CO_2 content and the associated increase in CH_4 content; ii) production of valuable biomass and iii) removal of nutrients from water and wastewater (Sutherland et al. 2019). In this process, the biogas generated from the anaerobic digester is fed into a photobioreactor where microalgae uptake CO_2 (Fig. 7) – a direct approach. This configuration was first introduced by Converti et al. (2009), who combined a mixed sludge anaerobic digester with a photobioreactor leading to biogas production with CH_4 content above 70%. Since then, higher methane content in the final biogas has been achieved with similar systems (Yan et al. 2013, Nagarajan et al. 2019, Bose et al. 2020). Yan et al. (2013) obtained a biomethane (92%

CH_4) through optimisation of culture conditions for the microalgae *Chlorella sp*. However, the research has identified several challenges that require future development for the emergence of this green technology (Nagarajan et al. 2019, Bose et al. 2020). The mass transfer and CO_2 solubility in the microalgal culture media is the first limitation (Bose et al. 2020). Unlike the water scrubbing process, gas is injected into the microalgal culture at atmospheric pressure and room temperature, limiting CO_2 solubility in the growth medium and leading up to 90% of input gas lost (de Godos et al. 2014). A second limitation is the high methane loss due to its solubility in a large volume of microalgal culture media. A third limitation is difficulty in harvesting the final biogas. If biogas is purged into the photobioreactor, an enclosed system is needed to collect the outlet gas. This requirement can limit the design for photobioreactors and microalgal growth. Another limitation is the lack of high CO_2 tolerant microalgal species. The high concentration of CO_2 in water reduces the pH value to below 6.0, which is detrimental to microalgal growth (i.e., disruption of cell membrane permeability and photosynthesis) (Sutherland et al. 2020). It is also a challenge because of the introduction of oxygen from the microalgal photosynthesis in to the final biogas.

Another approach to mitigate the limitation of the direct method is indirect biogas upgrading systems (Fig. 7). In this approach, CO_2 can be captured in a carbonate solution such as potassium carbonate. The potassium carbonate solution provides high quality of methane to be achieved. The saturated carbonate solution then is fed into the microalgal culture. Microalgae utilise bicarbonate as a carbon source for growth, regenerating the carbonate for a next biogas upgrading cycle. This approach, however; only limits to some specific microalgal species which can tolerate high ion strength and alkali environment (Xia et al. 2015).

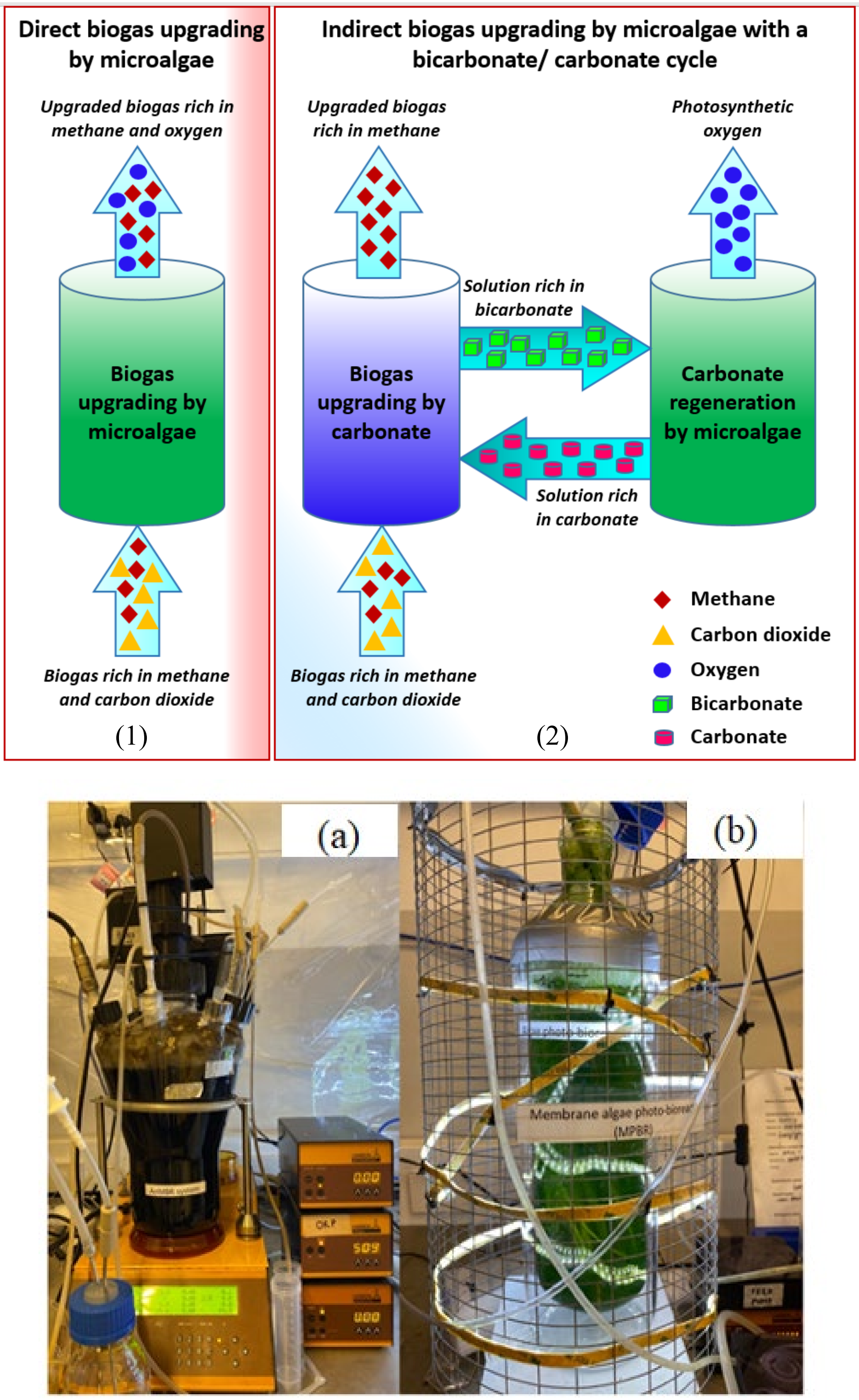

Figure 7: Schematic diagram of using microalgae for biogas upgrading (1) a direct and (2) indirect approach and a photography of anaerobic digester (a) coupled with a photobioreactor (b) for CO_2 capture and microalgae (*Chlorella vulgaris*) as an example of the direct approach

in our laboratories (Vu et al. 2020b). The schematic diagram was adapted from Xia et al. (2015).

7. Future perspectives

As AcoD continues to be adopted at WWTPs around the world, the demand for biogas upgrading technology to better utilise the surplus biomethane will continue to grow. Water scrubbing is currently the most widely applied technology due to low capital and operation cost. On the other hand, membrane separation has the highest growth. It is expected that membrane separation will overtake water scrubbing to become the most dominant technology for the biogas upgrading. It is also noteworthy that the technical readiness level for biogas upgrading is high with a variety of technologies that have been implemented at full-scale. In addition to water scrubbing and membrane separation, other technologies such as chemical scrubbing and pressure swing adsorption will continue to be utilised for biogas upgrading on a case to case basis.

Biogas upgrading to biomethane provides opportunities to tap into potential revenue that has not been previously utilised (IEA 2020). As discussed in section 3, raw biogas contains about 35% CO_2, which can be used for a range of applications. Gaseous CO_2 from the upgrading process can be used to produce dry ice at a temperature of – 78.5 °C. Unlike conventional ice, dry ice evaporates during the melting process, leaving no residue. Thus, dry ice is an appealing alternative to conventional ice in many industrial applications (e.g., food packing, biological samples transportation, and cleaning). The utilisation of this CO_2 source can provide additional revenue.

The quality of raw biogas could induce additional cost on overall expenditure of the pre-treatment and biogas upgrading processes. For example, high level of H_2S in raw biogas can increase the cost of its removal process (i.e. shorten the lifetime of adsorption column and increase chemical usage). Technologies to improve raw biogas quality and AcoD performance will have great benefits. To date, biological desulphurisation (i.e., injection air or oxygen into AcoD) to reduce H_2S formation has only been demonstrated in the laboratory. The variation in the performance between laboratory and full-scale studies may suggest more full-scale experience in the future. It is also recommended to evaluate the impact of air injection on other performance parameters (e.g., solid removal and biogas yields) and biosolid quality.

Biocatalytic enzyme and CO_2 capture by microalgae currently have a low technology readiness level. There is no study on the use of biocatalytic enzymes for biomethane production. It is also expected that the industrialized production of enzymes and its stability in the biomethane application is needed. Numerous questions need to be answered before deciding on an optimal microalgal biogas upgrading system. It is likely an innovative integrated system to i) use microalgae to capture CO_2; ii) to use anaerobic digestate as growth media and iii) to harvest microalgal biomass to use again as feedstocks for AcoD need to be evaluated in the upcoming studies.

8. Conclusion

Through anaerobic co-digesting sewage sludge and organic waste, numerous wastewater treatment plants (WWTPs) worldwide have achieved energy self-sufficiency and produced surplus biogas. Natural gas grid injection and transport fuels are attractive applications to utilise the surplus biogas from WWTPs after biogas upgrading to biomethane. Biogas upgrading technologies include water, organic and chemical scrubbing, pressure swing adsorption, membrane separation, and cryogenic are commercially available. Amongst them, water scrubbing is currently the most widely applied technology due to low capital and operation costs. On the other hand, the membrane separation is expected to be the dominant technology in the near future. In the 2015-2019 period, membrane process has a significant market growth (82% increase in new plants). Several emerging biotechnologies to improve biogas quality from co-digestion accelerate the absorption rate, and capture CO_2 in microalgal culture are highlighted and discussed. Information corroborated in this review demonstrates the possibility to transform WWTPs to net energy producers through the combination of co-digestion and biogas upgrade.

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