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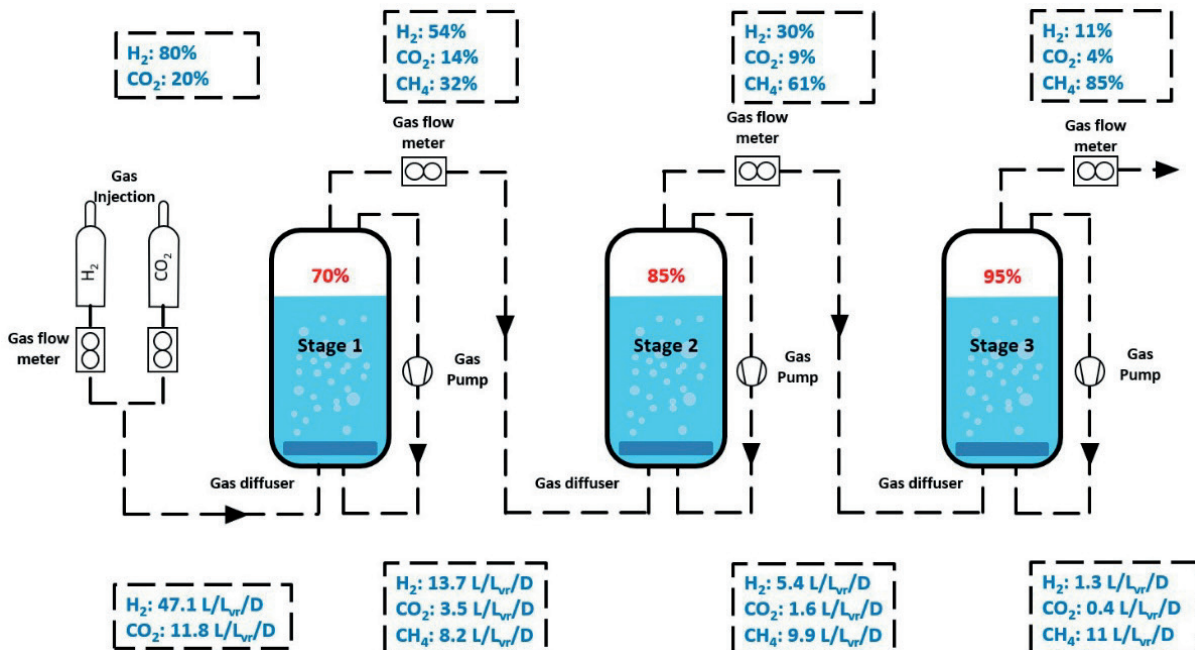
Technology Collaboration Programme

Circular economy approaches to integration of anaerobic digestion with Power to X technologies

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Circular economy approaches to integration of anaerobic digestion with Power to X technologies

Authors:

Jerry D Murphy, Davis Rusmanis, Nathan Gray, Richard O'Shea, MaREI centre, University College Cork, Ireland

Reviewed by

David Wall, John Paul Deane, and Maria Wellisch

Edited by

Jan Liebetrau

Cover graphic: Adapted from Voleklein et al. (2019)

Executive Summary

Many countries and geopolitical regions have ambitions of achieving net zero carbon by 2050 (US and EU) or 2060 (China). In advance of this, it is expected that a near zero carbon electricity system will be achieved in many jurisdictions around 2035. This will lend itself to electrification and associated decarbonisation of energy as much as is practicable; examples include battery electric vehicles and heat pumps for heating. However, in many countries it is seen that the future resource of renewable electricity (from wind and photovoltaic) will surpass the demand for electrification; hydrogen will be produced via water electrolysis to utilise larger resources of electricity or to facilitate electricity produced at times of low electrical demand. This renewable hydrogen will be used to displace existing fossil fuel sourced hydrogen used in industry, but beyond this renewable hydrogen will find new uses in the energy sector. Future markets for renewable hydrogen will include for direct use in transport such as in fuel cell vehicles for long-distance haulage. There may even be a limited market for hydrogen storage as a source of dispatchable electricity in periods where, for example, the wind profile decreases for long periods. The authors believe there will be a future role for renewable power to produce other energy carriers and chemicals. Generically known as 'Power to X' technologies, these technologies will produce renewable hydrogen molecules, which can be integrated with anaerobic digestion systems for production of methane (CH_4) and for production of methanol (CH_3OH: worldwide production of 110 million tonnes per annum). Power to X also includes for production of ammonia (NH_3) which has worldwide production of 160 million tonnes per annum.

One of the primary pathways of Power to X is power to methane ($4H_2 + CO_2 = CH_4 + 2H_2O$) whereby biogenic carbon dioxide (CO_2) is reacted with renewable hydrogen from water electrolysis using renewable electricity. A significant part of this report outlines the logistics of such systems when integrated with biogas facilities, including the narrative of a research laboratory in developing technologies that may be used as a biogas upgrading process. The starting point of the research lab was batch reaction of CO_2 and H_2 via hydrogenotrophic methanogens; the end point was a successful lab scale demonstration of a three-stage continuous ex-situ biological methanation rig. Several case stories are expanded upon including for demonstration and pre-commercial applications of catalytic and biological processes. An exemplar is detailed where an electrolyser and an ex-situ biomethanation system are situated within a wastewater treatment facility where oxygen generated during electrolysis may be valorised in the aeration basin. This in turn could reduce pumping of air by a factor of 5, and in so doing reducing the carbon footprint of the wastewater treatment facility by 40 %. The hydrogen can be used to upgrade the biogas from sludge digestion to biomethane suitable for compressed natural gas trucks, and the remaining hydrogen may be used to power fuel cells in heavy commercial trucks.

For net zero, decarbonisation must go beyond energy systems and extend to chemical manufacturing. Power to methanol is examined as a substitute for natural gas sourced methanol. Green methanol is seen as the route to decarbonising shipping. A future constraint will be limited resources of biogenic CO_2; fermentation and biomethane industries (with their by-product of concentrated cheap sustainable CO_2) can serve as a short-term solution of finite resource. A present source of low carbon methanol is steam reforming of biomethane; the present-day (2023) barriers to green ammonia production are the availability of significant resources of green hydrogen and the requirement for bunkering (and associated ship refuelling) in numerous strategic ports across the planet. Power to ammonia does not require biogenic CO_2 and can use the nitrogen that is resplendent in the air in the Haber-Bosch process. Green ammonia will be required to reduce the carbon footprints of fertiliser production and conventional agriculture.

This IEA Bioenergy Task 37 report employs an overarching perspective on circular economy approaches that could be used to integrate anaerobic digestion with Power to X technologies. It provides a menu of options that could be possible, whilst recognising that all solutions are bespoke. A significant benefit of renewable power to methane (and indeed power to X) is that it can build upon expensive existing infrastructure (such as the gas grid) and as such minimise total cost of ownership of new transport solutions. It can facilitate a practical transition to the net zero future.

This technical report is not a peer review paper but rather draws upon concepts within previous publications by IEA Bioenergy (from reports on broader topics) and other more recent peer reviewed scientific works, to provide a synthesised simplified specific bespoke narrative (in not overly technical language) of the rationale for application of Power to X technologies. The audience is assumed to be energy and infrastructure planners, technology developers, undergraduate students, utilities, and policy makers.

Table of Contents

1 Introduction

1.1 THE CHALLENGE OF DECARBONISING SOCIETY

The IPCC synthesis report (IPCC, 2014) suggested a carbon budget of 1000 Gt of CO_2 that could be emitted between 2011 and 2100 with a 66 % chance of not increasing global warming beyond 2 °C. An annual production of 40 Gt suggests that the budget would be used by 2035 if significant changes to industry and society are not made. Many geo-political regions have plans to achieve net zero greenhouse gas (GHG) emissions with deadlines ranging from 2050 (EU & US), 2060 (China) and 2070 (India).

The practicality of net zero emissions is extremely challenging. Major strides have been made in renewable electricity but less so in renewable heat (and cooling) and chemical manufacturing, and even less in transport. Decarbonising electricity and electrification of heat and transport is seen by many as a key pathway to net zero, but other pathways will be needed to abate the emissions from the very difficult to electrify (hard to abate) sectors of aviation, shipping, and long-distance haulage (Gray et al., 2021). Natural gas is a source of up to twice as much energy as the electricity grid in the US and the EU (Liebetrau et al., 2022). Many chemicals which we depend on (and produce in significant quantities) such as ammonia (NH_3) and methanol (CH_3OH) are sourced from natural gas. What will replace natural gas? What will be the source of the renewable hydrogen molecule for our fertilisers and plastics and all the chemicals and pharmaceuticals that our modern lives depend on?

There are elements of our existing economy that are very difficult to decarbonise. Infrastructure required to produce renewable electricity will require vast quantities of cement and steel which of themselves are very carbon intensive; we can use hydrogen to lessen the carbon footprint of steel but how do we decarbonise cement? A decarbonised society will also need to reduce the footprint of agriculture; is there a way to prevent cattle from belching methane and to prevent slurry storage sending fugitive methane emissions to the atmosphere? What about the carbon footprint of cheese and alcohol? Do we become vegetarian or vegan to approach net zero? What about waste and wastewater treatment facilities; these facilities will still emit methane even if all energy is electrified.

1.2 THE ROLE OF BIOGAS IN CIRCULAR ECONOMY SYSTEMS

IEA Bioenergy Task 37 has produced several documents that expound upon the role of biogas in a circular economy[1]. An anaerobic digester is not comparable to a wind turbine. A huge benefit of biogas is that it may be used for electricity, heat and/or transport, however it is much more than a source of energy (Liebetrau et al., 2021). Anaerobic digestion is a means of reducing fugitive methane emissions in slurry storage (Liebetrau et al., 2017) or waste management. It is part of a system that is used in waste management to minimise the environmental impact of municipal or industrial wet organic waste. Anaerobic digestion systems can play an essential role in managing water pollution and in smart agriculture (McCabe et al., 2020). The by-products of anaerobic digestion include for biofertilizer which reduces the need for fossil fuel-based fertiliser (sourced from NH_3, which in turn is sourced from natural gas); indeed, anaerobic digestion can play an essential role in organic farming converting dung to liquid biofertiliser through mineralisation of nutrients (IEA Bioenergy, 2019).

CO_2 is a by-product of anaerobic digestion that can be critical in assessing sustainability of a biogas system in terms of carbon footprint (gCO_2 per MJ of energy vector). If biogas is segregated into biomethane (for gas grid injection or transport fuel) the remaining gas stream (almost pure CO_2) should not be discharged to the atmosphere but rather be captured. This is the realm of bioenergy with carbon capture and sequestration (BECCS); in a BECCS system CO_2 could be sequestered and stored underground and the whole system would be a negative emission technology (EASAC, 2018). However, carbon sequestration in geological strata is not a mature technology and very small quantities of CO_2 are stored in this fashion. Bioenergy with carbon capture and utilisation is more readily applicable in 2023.

There are many uses for CO_2; the Danish market for CO_2 is of the order of 65,000 tonnes per annum

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[1] https://task37.ieabioenergy.com

and the uses include for laboratories, hospitals, packaging of meat, slaughterhouses, carbonation of beer and soft drinks (IEA Bioenergy, 2020). It is expected in future decarbonisation pathways, that renewable hydrocarbons will be required for the hard to abate sectors of aviation, shipping, and long-distance haulage. CO_2 segregated from biomethane from biogas upgrading, will be a concentrated, cheap, biogenic, and renewable source of CO_2. The resource of this biogenic CO_2 is essential for Power to X to produce renewable fuels (or electrofuels) and chemicals. RePowerEU, (2022) proposes a 10-fold increase in biomethane within the EU, from 3.6 billion cubic meters in 2021 to 35 billion cubic meters in 2030. For example, the Climate Action Plan of Ireland (an EU member state with a population of about 5 million) mandates 5.7 TWh of biomethane by 2030 (Rialtas na hÉireann, 2022). This is equivalent to 650 MW of installed capacity or 200 number 3.25 MW biomethane facilities. The by-product (or co-product) of biomethane creates a significant renewable CO_2 resource at over 98 % concentration.

1.3 RATIONALE FOR POWER TO X

The future of decarbonisation will lead to significant increases in commercial availability of renewable electricity. For example, the nine countries of the North Seas Energy Cooperation (Belgium, Denmark, France, Germany, Ireland, Luxembourg, the Netherlands, Norway, and Sweden) together with the European Commission, have agreed on a target of 260 GW of offshore wind energy by 2050[2].

We may use as an example, Ireland in the northeast Atlantic which has achieved 36.4 % renewable energy supply in electricity (RES-E) in 2021[3]. Ireland has a target of 5 GW of offshore wind-sourced renewable electricity and a further 2 GW of hydrogen from offshore wind by 2030[4]. This is proposed to lead to 80 % RES-E by 2030. Temporal mismatches such as windy summer nights with low electricity demand will lead to periods of oversupply; this typically would lead to dispatch down through curtailment. It is proposed that in future years this "oversupply" will be used to make hydrogen via electrolysis. The resource of offshore wind beyond 2030 is estimated by industry to be of the order of 70 GW[5]; this is significantly more than required for the indigenous supply of electricity or indeed energy that may be electrified. This is where electricity may be used to produce Power to X products in the form of electro-fuels or renewable chemicals.

1.4 POWER TO HYDROGEN

The first step in Power to X is the production of hydrogen via electrolysis of water; whereby direct electrical current (DC) is used to split water into oxygen and hydrogen as per equation 1.1.

$$2 H_2O \text{ (liquid)} = \quad 2 H_2 \text{ (gas)} + O_2 \text{ (gas)} \qquad \text{Eq. 1.1}$$

The electrolyser has two electrodes (an anode and a cathode), an electrolyte and a membrane which hinders recombination of the hydrogen and oxygen gas. Hydrogen is formed at the cathode and oxygen at the anode. The reaction is endothermic and energy input is required in the form of electricity. There are three electrolysis technologies: Alkaline Electrolysis Cells (AEC); Proton Exchange Membrane (PEM) cells; and Solid Oxide Electrolysis Cells (SOEC). One of the issues with hydrogen as an energy vector is its volumetric energy density. Though it has a high energy content per unit mass (120 MJ/kg or 33.33 kWh/kg) because its density is very low (0.09 kg/m³ at 0 °C and 1 bar) it has a volumetric energy density of 10.8 MJ/m³ or 3 kWh/m³. As such hydrogen is difficult to store and transport (without even taking into consideration explosive hazards). This leads us to further vectors in Power to X; typically, the second step is power to methane.

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[2] https://www.gov.ie/en/press-release/9c152-members-of-the-north-seas-energy-cooperation-nsec-grasp-historic-opportunity-to-accelerate-europes-move-towards-energy-independence/

[3] https://www.seai.ie/data-and-insights/seai-statistics/key-statistics/renewables/

[4] https://www.gov.ie/en/publication/f3bb6-policy-statement-on-the-framework-for-phase-two-offshore-wind/

[5] https://www.greenrebel.ie/wind-energy-irelands-renewables-in-ireland-2022/

1.5 POWER TO METHANE

It may be asked why it would make sense to convert electricity to hydrogen first, and then to methane. There are many combined cycle gas turbines (CCGT) which convert natural gas (methane) to electricity (methane to power). So, why reverse the process and convert power to methane?

The rationale for power to methane is in the broader energy realm and indeed beyond energy as a feedstock for the chemical industry. The natural gas grid is a source of more energy than the electricity grid in the EU and the US. Natural gas is a significant source of power, and a lot of industries are optimised for use of natural gas as an energy vector. Many facilities in the food and beverage sector are set up for use of natural gas as a source of process heat to evaporate alcohol off stillage, or segregate milk solids from water; a change to electricity, if plausible, could necessitate significant investment in electrical infrastructure, often in relatively rural locations. For some industries, such as glass production, a gaseous fuel vector is essential. Beyond energy, methane (CH_4) has been an ingredient in ammonia (NH_3), methanol (CH_3OH) and other chemical synthesis processes.

As in the energy transition, a substitute for natural gas (fossil methane) is highly desirable, if not essential. In decarbonising society, we need renewable molecules and in particular renewable methane and/or hydrogen molecules. Such renewable hydrogen molecules may be sourced from biomethane (CH_4) or from H_2 via water electrolysis (which as previously discussed has some issues with volumetric energy density) or may be in the form of synthetic methane derived from hydrogen (Power to X).

The reasons for converting hydrogen to methane in Power to X pathways are threefold. A major rationale is infrastructure: the existence of the natural gas grid; existing Combined Cycle Gas Turbine (CCGT) power plants; and the facilities in place at industry level that are currently based on natural gas and methane. It could be said it is the path of least resistance in that all the distribution systems are in place, and we just need to decarbonise the fuel, in this case methane. The second reason has to do with the gas itself. Methane has a volumetric energy density of 10 kWh/m³ which is over 3 times more energy dense than hydrogen at 3 kWh/m³. The third reason is the explosive risk associated with hydrogen. A recent risk assessment conducted by Arup on behalf of the UK Government[6] suggested that using hydrogen in the home, instead of natural gas, would lead to an increase in domestic gas explosions by a factor of 4.

The power to methane process as outlined in Equation 1.2 requires a source of CO_2. Utilisation of fossil fuel sourced CO_2 would not be considered sustainable as it would support the continued on-going use of fossil fuels, which are preferably kept in the ground. This brings us to biogenic CO_2 produced with as little fossil fuel input as possible. Segregation of biogas into biomethane and CO_2 leads to a concentrated and cheap source of CO_2.

$$4H_2 + CO_2 \quad = \quad CH_4 \text{ (gas)} + 2H_2O \text{ (gas)} \qquad\qquad \text{Eq. 1.2}$$
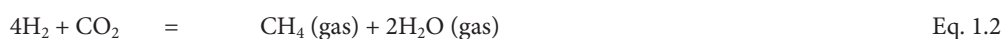
1.6 BIOGENIC CO_2

When we examine true biogenic sources of CO_2, they are relatively limited. Fermentation within alcohol and ethanol production release relatively pure CO_2. However, if we assess the overall brewing and distillation process not all the released CO_2 is biogenic; if for example, we capture CO_2 from the chimney stack of a brewery or distillery, although the fermentation may be considered a biological process (conversion of grains to alcohol), typically natural gas would be used to provide process heat to the evaporation process and the CO_2 from the exhaust stack would not be biogenic. For the CO_2 from the exhaust stack to be fully biogenic, the brewery or distillery would need to operate on biomethane or a green gas, not natural gas. In a circular economy system, this would involve digesting the by-products (such as stillage and draff) from the distillery, producing biogas and using it to replace natural gas in the alcohol production process; this would reduce scope 1 emissions of the distillery (O'Shea et al., 2022). Application of the digestate to agricultural land that produced crops for the distillery would reduce scope 3 emissions of the

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[6] https://www.rechargenews.com/energy-transition/hydrogen-in-the-home-would-be-four-times-more-dangerous-than-natural-gas-government-report/2-1-1047218

distillery (and scope 1 emissions of the farm where the crops were grown). As such to decarbonise a distillery, the use of biomethane within the process has an added benefit in that the CO_2 emitted in the stack can, if captured, be a source of biogenic carbon for Power to X applications.

Another approach would be to capture the CO_2 from a biomethane facility. This can be configured in many ways, and it is plausible that addition of hydrogen to biogas may replace the biogas upgrading process. If we were to consider that biogas consists of 60 % CH_4 and 40 % CO_2, then reaction of the hydrogen (from an electrolyser) with the CO_2 in the biogas would result in conversion of CO_2 to methane with an overall increase in methane output of the biogas system of typically 67 % (40 % CO_2 converted to CH_4 expressed as a percentage of the initial 60 % CH_4). As such the power to methane system would replace traditional biogas upgrading systems, increase the yield of methane and in so doing, optimise the economics of power to methane. The resource of this CO_2 is however limited when we think of the resource required to fuel ships and planes. This brings us to direct air carbon capture which theoretically is abundant in supply and would utilise renewable electricity to power such a process. The cost may be expensive, but the process may be essential to effect net zero.

1.7 RENEWABLE HYDROCARBONS AND RENEWABLE AMMONIA

In examining shipping and aviation, there is little potential for electrification; batteries (as the technology stands in 2023) require too much space and weigh too much (Gray et al., 2021). We need fuels with a high energy density. This is also problematic for power to gas. If we consider diesel at 10 kWh per litre (kWh/L) then methane (10 kWh/m³) and hydrogen (3 kWh/m³) are 1000 and 3000 times more voluminous per unit of energy at standard temperature and pressure. Typically, methane is compressed to 250 bar and hydrogen to 800 bar, leading to approximate volumetric energy densities of approximately 2.5 kWh/L for compressed methane and 2.4 kWh/L for compressed hydrogen. These gaseous fuels would still require 4 times the storage capacity of diesel for the same energy stored or else fuel a vehicle for 25 % of the distance based on the same storage volume.

When we look at low carbon liquid fuels, we tend to consider biofuels. Ethanol (21 MJ/L or 5.8 kWh/L) and biodiesel (fatty acid methyl ester at 34 MJ/L or 9.4 kWh/L) have a role to play in terms of private cars and fleets of light goods vehicles. However, the resource is limited in comparison to the resource required for aviation, shipping, and long-distance haulage; there are also concerns about the sustainability of energy crops, especially as the required resource increases.

This leads us to power to liquids, such as methanol and ammonia. Worldwide production of methanol is of the order of 110 million tonnes. It is expected to be a major source of shipping fuel which will increase worldwide production. Presently, it is made from natural gas and in a decarbonised world it will be a major demand source for hydrogen from renewable electricity and biogenic CO_2. Methanol (CH_3OH) has a similar stoichiometry to methane (CH_4) and with reference to Equation 1.3, it may be seen how we can convert biogas to methanol. Of issue here is the requirement for a biogas with a composition of 75 % CH_4 and 25 % CO_2. This may be overcome by limiting the CO_2 to 25 % of the overall biogas in the process or reacting hydrogen from electrolysis with CO (generated by splitting biogas into CO and H_2 using an electrically driven catalytic converter). Methanol synthesis as per equation 1.4 is an exothermic reaction conducted between 200 and 300 °C and 3.5 to 10 MPa (IEA Bioenergy, 2020b).

$$3CH_4 + CO_2 + 2H_2O \quad = \quad 4CH_3OH \qquad\qquad\qquad \text{Eq. 1.3}$$
$$CO + 2H_2 \quad = \quad CH_3OH \qquad\qquad\qquad\qquad \text{Eq. 1.4}$$

Another energy vector is ammonia. Worldwide production of ammonia is of the order of 160 million tonnes and is the route to fertiliser. A major benefit of power to ammonia is that we are not constrained by biogenic CO_2 but rather can use nitrogen from the atmosphere as per Equation 1.5.

$$N_2 + 3H_2 \quad = \quad 2NH_3 \qquad\qquad\qquad\qquad\qquad \text{Eq. 1.5}$$

1.8 BRIEF OF REPORT

This report aims to provide an overview of the role that anaerobic digestion can play in Power to X technologies. This is not a peer review paper but rather aims to provide an understanding to readers from an audience of technology developers, energy and infrastructure planners, undergraduate students, utilities, and policy makers. The objectives are to:

- Outline processes which integrate anaerobic digestion with electrolysis (Chapter 2);
- Detail the design evolution of an experimental laboratory biomethanation process (Chapter 3);
- Examine real world circular economy applications of integration of anaerobic digestion with power to methane systems (Chapter 4);
- Assess future applications integrating anaerobic digestion with Power to X technologies such as production of methanol and ammonia (Chapter 5);
- Discuss optimal routes and applications for Power to X technologies (Chapter 6).

2 Integration of anaerobic digestion with electrolysis

In this chapter, we consider types of electrolysers and their ability to react to the intermittency of variable renewable electricity. We further examine biomethanation technologies (both catalytic and biological) and assess how these technologies can be combined to best effect at an anaerobic digestion facility.

2.1 HYDROGEN PRODUCTION VIA ELECTROLYSIS

For a more detailed description of electrolysers, the reader is referred to section 4.1 of our previous IEA Bioenergy Task 37 report (Person et al., 2014) "A perspective on the potential role of biogas in smart energy grids" and section 3 of our previous IEA Bioenergy Task 37 report (Liebetrau et al., 2020) "Integration of biogas systems into the energy system: Technical aspects of flexible plant operation." In synthesis, there are three electrolysis technologies: Alkaline Electrolysis Cells (AEC); Proton Exchange Membrane (PEM) cells; and Solid Oxide Electrolysis Cells (SOEC). Of these, the AEC is the most developed; the PEM is more suited to variable renewable electricity due to its more flexible operation and the SOEC is the least mature but offers higher electrical efficiency if there is a source of available thermal energy (Table 2.1). For the developed electrolysers (AEC and PEM) the efficiencies of conversion of electricity to hydrogen are optimistically about 70 %; these operate at pressures in the range of 10 to 50 bar and at temperatures of 10 to 90 °C. PEM has the significant advantage of quick response and minimal cold start, which is crucial for systems utilising variable renewable electricity.

Table 2.1: Selected technical characteristics for different electrolysers in a Power to Gas system in 2020 (Liebetrau et al., 2020 which is adapted from Buttler & Spliethoff, 2018, Schmidt et al., 2017, and McDonagh et al., 2018)

Electrolyser type	Alkaline Electrolysis Cell	Proton Electrolyte Membrane	Solid Oxide Electrolysis Cell
Operating cell temperature (°C)	60 - 90	50 - 80	700 - 900
Operating pressure (bar)	10 - 30	20 - 50	1 - 15
Hydrogen production rate (m^3 H_2/h)	< 1400	< 400	< 10
System energy consumption (kWh/m^3 H_2)	4.3 - 5.4	4.3 - 5.3	3.9 - 4.4
Process efficiency (%)[1]	65 - 82	67 - 82	80 - 90
Cold start time (min)	60 - 120	5 - 10	hours
Maturity of technology	Mature	Commercial	Demonstration

[1] The process efficiency may be evaluated based on dividing the HHV of H_2 (3.54 kWh/m^3) by the system energy consumption; for example, for AEC this may be evaluated in the range (3.54/5.4 =) 65 % to 82 % (3.54/4.3)

2.2 CATALYTIC CONVERSION OF HYDROGEN TO METHANE

Biogas may be upgraded in a power to methane process to almost pure methane. Alternatively, the biogas may be upgraded by traditional physio-chemical processes and subsequently the CO_2 (segregated from the biomethane stream) may be reacted with hydrogen (from electrolysis) to produce another methane product (which may be termed synthetic methane). The power to methane process can be a catalytic process or a biological process. Catalytic methanation employs nickel (or ruthenium based) catalysts and operates within a temperature range of 300 to 500 °C. The catalytic process requires that hydrogen sulphide (H_2S) and siloxanes must be removed in advance of the catalytic step.

Carbon monoxide, carbon dioxide and hydrogen may be converted to methane and water by the processes outlined in Equations 2.1 and 2.2 (ΔHr at 25 °C). Equation 2.1 is known as the Sabatier Process.

$$4H_2 + CO_2 \quad = \quad CH_4 + 2H_2O \qquad \Delta Hr = \text{-165 kJ mol-1} \qquad \text{Eq. 2.1}$$
$$3H_2 + CO \quad = \quad CH_4 + H_2O \qquad \Delta Hr = \text{-210 kJ mol-1} \qquad \text{Eq. 2.2}$$

These reactions are thermodynamically favourable and generate significant quantities of heat (Persson et al., 2014). As such this heat may be used, for example within a district heating system. One circular economy process that may be employed is to use the exothermic heat from catalytic conversion to provide the heat input to a Solid Oxide Electrolysis Cell and to avail of the improved electrical conversion to hydrogen (c. 85 % refer table 2.1).

If we consider the Sabatier reaction it may be noted that 4 moles of hydrogen are required to produce 1 mole of methane, thus it takes 12 kWh (4 * 3kWh/m³ H_2) to produce 10 kWh (10 kWh/m³ CH_4); a maximum theoretical efficiency of 83 %. The actual efficiency is expected at approximately 80 % according to Benjaminsson et al. (2013). The reader is referred to section 4.3 of our IEA bioenergy publication (Persson et al., 2014) for more detail on the catalytic power to methane process.

2.3 BIOLOGICAL CONVERSION OF HYDROGEN TO METHANE IN A BATCH SYSTEM

The application of catalytic processes operating at 300 to 500 °C may be too high a specification for farm scale biogas plants. This report is more focused on biological processes; the utilisation of an anaerobic biological process at a biogas facility has significant benefits. The anaerobic process is well documented and may be greatly simplified as in Figure 2.1. In brief wet organic feedstock is hydrolysed to sugars (by hydrolytic bacteria) which in turn are fermented to volatile fatty acids (VFAs) by fermentative bacteria. These volatile fatty acids may be further converted by acetogenic bacteria to acetate (CH_3COO^-), hydrogen, and carbon dioxide. If for example, we examine the breakdown of ethanol (CH_3CH_2OH) as a representative volatile fatty acid, species 2 the acetogenic bacteria produce hydrogen and carbon dioxide while the hydrogenotrophic methanogenic archaea (species 4.1) use hydrogen and carbon dioxide as ingredients to produce methane within a favourable exothermic reaction. For biomethanation we are most interested in species 4.1 (the hydrogenotrophic methanogenic archaea).
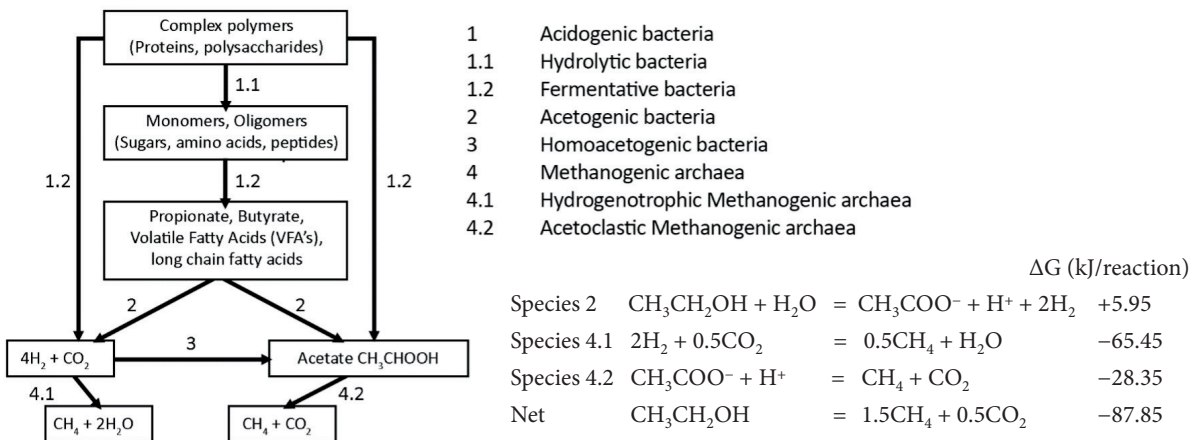

1	Acidogenic bacteria
1.1	Hydrolytic bacteria
1.2	Fermentative bacteria
2	Acetogenic bacteria
3	Homoacetogenic bacteria
4	Methanogenic archaea
4.1	Hydrogenotrophic Methanogenic archaea
4.2	Acetoclastic Methanogenic archaea

ΔG (kJ/reaction)

Species 2	$CH_3CH_2OH + H_2O$	$=$	$CH_3COO^- + H^+ + 2H_2$	+5.95
Species 4.1	$2H_2 + 0.5CO_2$	$=$	$0.5CH_4 + H_2O$	−65.45
Species 4.2	$CH_3COO^- + H^+$	$=$	$CH_4 + CO_2$	−28.35
Net	CH_3CH_2OH	$=$	$1.5CH_4 + 0.5CO_2$	−87.85

Figure 2.1. Four trophic groups involved in anaerobic digestion (adapted from Persson et al., 2014)

2.4 POSSIBLE CONFIGURATIONS OF POWER TO METHANE SYSTEMS

Several configurations that may be used to employ a power to methane process at a biogas facility. In Figure 2.2A hydrogen from an electrolyser is added to an existing anaerobic reactor. This would be considered *an in-situ biological methanation* process. With reference to figure 2.1 acetoclastic methanogenic archaea would continue to produce CO_2 and minimise potential to reach a biomethane standard (with methane content more than 97 %). Another issue is the rise in the hydrogen partial pressure within the digester. In assessing the stoichiometry associated with species 2 (in Figure 2.1) it may be noted that the Gibbs Free Energy is positive and as such the reaction is not favourable. This may be explained with reference to the hydrogen partial pressure. The reason the anaerobic process works is the syntrophic mu-

tualism that exists between acetogenic bacteria (species 2) and hydrogenotrophic methanogenic archaea (species 4.1). Hydrogenotrophic methanogenic archaea have a high affinity for hydrogen as shown by the negative Gibbs Free Energy and as such the archaea efficiently remove the hydrogen produced by the acetogenic bacteria (Figure 2.1) which (following Le Chatelier's Principle[7]) reduces hydrogen partial pressure and facilitates the reaction. As such this configuration would need to be followed by a traditional biogas upgrading step to achieve a biomethane standard.

In Figure 2.2B the biogas and the hydrogen are reacted together in an *ex-situ biological methanation* process. In theory this configuration can replace traditional biogas upgrading processes and as such act as a biogas upgrading process. This is a huge benefit as the CAPEX and OPEX of traditional physio-chemical biogas upgrading can be avoided thus reducing the overall costs substantially. Of potential issue, however, is that the methane that accompanies the CO_2 within the biogas was shown by Voelklein et al., (2019) to impact negatively on the efficiency of the process due to the impact of the presence of methane on the partial pressure of the other gases.

In Figure 2.2C biogas is upgraded to biomethane, and the concentrated form of CO_2 is reacted with the hydrogen to produce a second stream of synthetic methane. It was shown by Voelklein et al. (2019) that this is a more efficient process, however a negative aspect of this, is that the developer must also invest in traditional biogas upgrading raising the costs of the process.
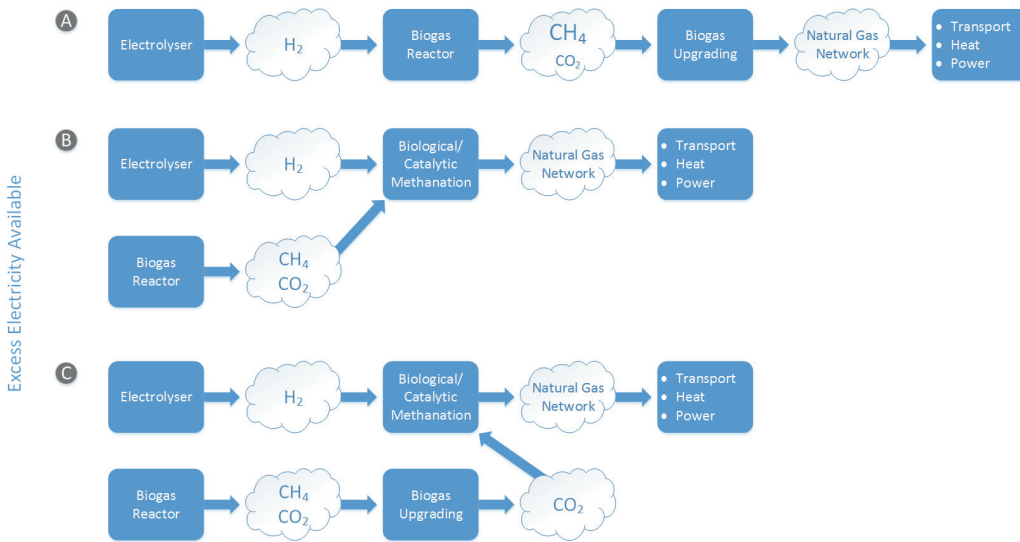

Figure 2.2. Different pathways that can be used to integrate an electrolyser at a biogas facility (from Persson et al., 2014)

2.5 CHAPTER SYNTHESIS

In our future decarbonised society, it is likely that renewable electricity will provide more electricity than is required for electrification. Hydrogen may be generated from renewable electricity but what will the hydrogen be used for? A pathway to convert this hydrogen to a renewable hydrocarbon is to react with biogenic CO_2. The most abundant source of high concentration biogenic CO_2 may be from biomethane systems. As such there is a significant benefit in integrating anaerobic digestion with electrolysis. The reaction process may be carried out catalytically, but the authors see biological methanation as more synergistic to anaerobic digestion. Biological methanation effected by hydrogenotrophic methanogenic archaea is a technology that may substitute for traditional biogas upgrading. The next chapter will assess in detail how the technology works through a narrative of development over the years in a research laboratory.

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[7] Le ChatElier's Principle may be synthesised for this case as follows "If a chemical reaction is at equilibrium and experiences a change in concentration of products, the equilibrium shifts in the opposite direction to offset the change."

3 Design and output of experimental laboratory biomethanation process

The concept of this chapter is to deepen the understanding of the reader in the science and engineering of power to methane technologies. This is effected through the narrative of one laboratory's journey in researching conversion of CO_2 to CH_4 starting from a simple batch laboratory system and working up to a lab scale demonstration system with each step increasing the technology readiness level (TRL). This is not meant to be exhaustive, nor deep fundamental science, but rather a synthesis of pieces of research required to develop a particular bespoke technology. There are many other technologies; this is but one such example. *The non-technical reader who may be overawed by the scientific and engineering detail may skip to Chapter 4 without impacting on the overall story within this report.*

3.1 BATCH BIOLOGICAL METHANATION PROCESS

For biological methanation we are most interested in hydrogenotrophic methanogenic archaea (See species 4.1 in Figure 2.1). Guneratnam et al. (2017) undertook a basic study of batch ex-situ biological methanation and found that in feeding carbon dioxide and hydrogen to a mixed culture of anaerobic microbes at 65 °C (Figure 3.1) that a gas with a methane composition of 92 % was produced within a 24 hour period. *Methanothermobacter wolfeii* was dominant in the microbial community: *M. wolfeii* is known to thrive between 55 and 65 °C in a pH range of 7.2 to 7.6. In this batch process the volumetric productivity was reported as 0.45 L CH_4/L reactor per day (Guneratnam et al., 2017).

Figure 3.1. Simple experimental examination of ex-situ biological methanation (from Guneratnam et al., 2017)

3.2 MODEL OF A CONTINUOUS EX SITU BIOLOGICAL METHANATION PROCESS

Voelklein et al., (2019) continued this work at a larger scale with an ambition of increasing the technology readiness level. They used a series of experimental processes on a single stainless steel 9.5 L reactor operating in a continuous ex-situ biomethanation mode at thermophilic temperatures (55 °C). The input gas was defined; based on Equation 2.1 ($4H_2 + CO_2 = CH_4 + 2H_2O$) the stoichiometric ratio of H_2 to CO_2 in the initial trial was set at 4:1. The output of this first trial (54 % H_2,14 % CO_2 & 32 % CH_4) was generated as the input for the second trial on the same reactor. The output of the second experimental process (30 % H_2, 9 % CO_2 & 61 % CH_4) became the input for the third trial. The final quality achieved was (11 % H_2, 4 % CO_2 & 85 % CH_4) The system described in Figure 3.2 was modelled based on these series of experiment trials on the one 9.5 L stainless steel reactor. The modelled system may be described as a cascading system with recirculation of the gas via a ceramic diffuser to enhance the dissolution of hydrogen within the microbial broth. The limited solubility of hydrogen is suggested as a limitation of such systems. Use of high pressures and diffusers is one means of overcoming this limitation.
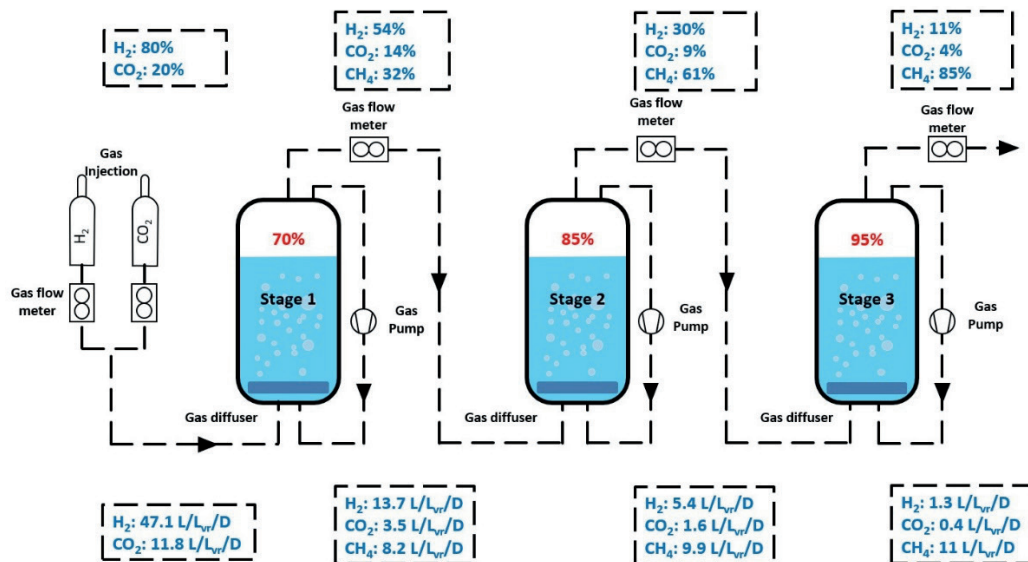
Figure 3.2. Experimental and modelled output of a small lab scale continuous ex-situ biological methanation process (adapted from Voelklein et al., 2019)

3.3 SET UP OF A LABORATORY CONTINUOUS EX SITU BIOLOGICAL METHANATION PROCESS

To take the model outlined above to a higher technology readiness level, a laboratory-based demonstration system was designed and commissioned as per Figure 3.3. The concept here was to take the work of Voelklein et al. (2019), which was based on one reactor and construct three reactors so that the output from the first would flow directly to the second and on to the third in one continuous sweep. In essence the overarching ambition was to operate the modelled system at a scale bigger than lab scale and in a continuous mode. This involved an increase in the technology readiness level of this bespoke designed system.
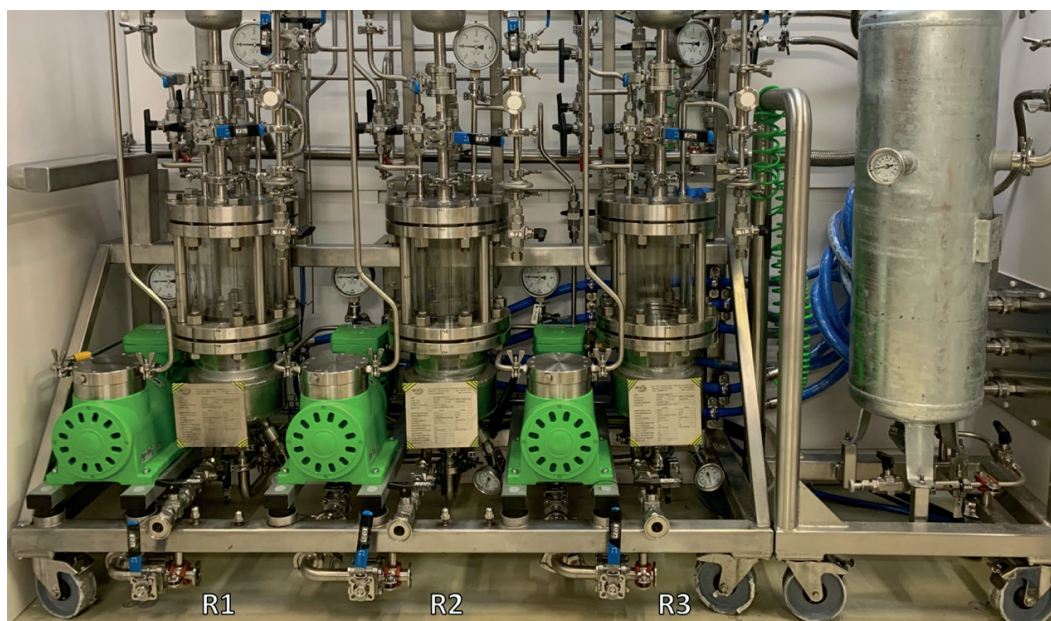

Figure 3.3. Demonstration scale continuous ex-situ biological methanation rig

The experimental process employed individual gas supply lines for CH_4, H_2, CO_2 and N_2 connected to an external manifold. Each gas line had an upstream pressure of 5 barG, with manually operated rotameters controlling the flow. These lines merge into the manifold mixing tank to ensure mixing of gases before entering the contiguous reactor system. The piping used was 316 L high-grade stainless steel to eliminate

leaks from the pipeline joints. The nitrogen supply was controlled via a simple pressure indication and control valve and its function was to render the internal system environment inert when required. The inlet flow rotameters for hydrogen, carbon dioxide and methane had a varied operational range: H_2 flow rates of 4.1 to $39.0 \, L_N/h$ (litres per hour at standard temperature and pressure); CO_2 flow rates of 0.9 to $9.0 \, L_N/h$; and CH_4 flow rates of 1.7 to $17.0 \, L_N/h$. These ratios reflect the 4:1 stoichiometric ratio of H_2 to CO_2 in the Sabatier reaction. The ratio between CO_2 and CH_4 is reflective of a composition of biogas of about 65 % CH_4 and 35 % CO_2.

Three identical 6L (working volume) reactor chambers of 150 mm diameter constructed from 316L stainless steel were used in the experimental process (see Figure 3.4). The lower steel basin housed a custom diffuser plate ("B" in Figure 3.4) and a liquid take-off valve (C in Figure 3.4). The diffusers provide $140 \, cm^2$ of diffuser area which generate bubbles of size less than 1 mm; the diffuser can deal with a maximum flow rate of 9 normal litres per minute (L_n/min) or $540 \, L_n/h$. The upper sight glass section is made of borosilicate glass, which allows for observation of the reactor during operation. The reactors include for the flat top flange, holding the feed-in valves, sealing the reactor and gas recirculation/outlet lines. Between the components, a polytetrafluoroethylene (PTFE) gasket ensures a gas-tight seal at the flange. Double rubber O-rings between the flange and the borosilicate glass seal the interface between the glass and steel. The installed pressure safety valves (PSV) are set to 2.5 barG, to prevent excess pressure build-up should the outlet gas flow be restricted at any point in the system.
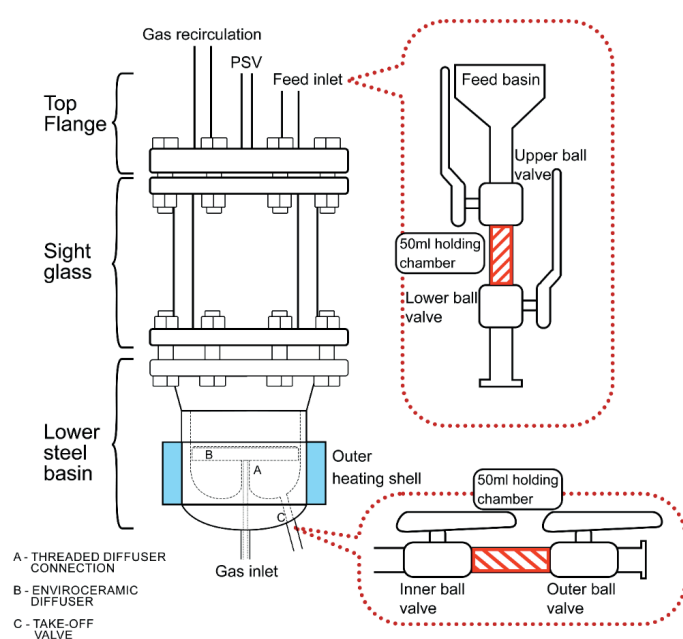
Figure 3.4: Profile of a methanation reactor chamber, where (A) is the threaded diffuser connection, (B) is the diffuser and (C) is the liquid take-off line. PSV – Pressure safety valve.

A methanogen-rich liquor was harvested from a thermophilic laboratory anaerobic digester processing grass silage. The liquor was sieved via a 150 μm mesh to remove particulates, leaving solely the liquor component to be added to the biological methanation rig. The resulting inoculum had a total solids (TS) content of 1.5 %, a volatile solids (VS) content of 0.9 % and an initial pH of 7.40. Over time, with feed only of hydrogen and carbon dioxide gases, the inoculum became dominated with hydrogenotrophic methanogenic archaea.

Nutrients were added via an airlock system (comprising two ball valves and a funnel) facilitating addition of liquids without allowing air in or gases out. Similarly, the outlet airlock system allowed for the extraction of liquors for sampling (Figure 3.4 C).

3.4 ANALYSIS OF EX SITU BIOLOGICAL METHANATION PROCESS

The measurement of gases was normalised to a dry gas at standard temperature and pressure (273.15 K, 1 atm). Methane evolution rate (MER) is used as a metric to allow comparison of systems. It may be defined as per Equation 3.1.

$$MER = \frac{Q_{CH4,out} - Q_{CH4,in}}{V_R} \qquad \text{Eq. 3.1}$$

Where, MER is the methane evolution rate measured in L CH_4 per litre reactor per day (L/L_{vr}/d), $Q_{CH4,out}$ is the flow rate of CH_4 out of the system, Q_{CH4in} is the flow rate of CH_4 into the system and Vr is the reactor volume (L).

To measure the efficiency of the process the process was monitored for CO_2 conversion efficiency as per Equation 3.2

$$X_{CO_2} = \frac{\%_{CH4}}{\%_{CO_2} + \%_{CH4}} \qquad \text{Eq. 3.2}$$

Where X_{CO2} is the carbon conversion efficiency, $\%_{CH4}$ is the outlet methane composition and $\%_{CO2}$ is the outlet carbon dioxide composition.

3.5 OUTPUT OF EX SITU BIOLOGICAL METHANATION SYSTEM

The design of the experiment was split into three stages.

Stage 1: For the first 20 days a volumetric throughput of hydrogen of 34.6 L per litre reactor per day was used (Table 3.1 and Figure 3.5). The corresponding CO_2 throughput was 9.4 L per litre reactor per day equivalent to a stoichiometric ratio of H_2 to CO_2 of 3.7:1; the rotameter sensitivity made it very difficult to get an addition of an exact 4:1 ratio. This was a successful biological process with CO_2 conversion efficiencies of 72 % in Reactor 1 (R1), 92 % in Reactor 2 (R2) and 97 % in Reactor 3 (R3). The average methane evolution rate was evaluated as 2.9 L CH_4 per litre reactor per day.

Stage 2: The throughput of gases were raised to 50.9 L_{H2}/L_{VR}/d and 11.8 L_{CO2}/L_{VR}/d equivalent to a stoichiometric ratio of 4.3:1 (H_2:CO_2). With reference to Figure 3.5 this proved very challenging for the biological system with methane composition dropping very significantly across the three reactors, especially reactor 1 and 2.

Stage 3: From day 35 on the throughput of gases were reduced to a similar level to stage 1; 35.6 LH_2/L_{VR}/d and 9.7 L_{CO2}/L_{VR}/d equivalent to a stoichiometric ratio of 3.6 (H_2:CO_2). The system came back to a high biological efficiency with methane compositions more than 90%.

It should be noted that gas volume will reduce as voluminous hydrogen (density of 0.089kg/m³) is converted to more dense CH_4 (density of 0.713 kg/m³) resulting in lower volumes of gas in output than input.

Table 3.1: Output of experimental process for Stages 1, 2, and 3 of this study as compared to values from Voeklein at al., 2019.

Study	Units	Voelklein et al.	Stage 1	Stage 2	Stage 3
H_2	L/L_{vr}/d	47.1	34.6	50.9	35.6
CO_2	L/L_{vr}/d	11.8	9.4	11.8	9.7
H_2:CO_2		4.0	3.7	4.3	3.6
MER Reactor 1	L/L_{vr}/d	8.2	6.3	3.1	4.5
MER Reactor 2	L/L_{vr}/d	1.7	2.1	4.0	2.9
MER Reactor 3	L/L_{vr}/d	0.85	0.3	3.2	0.2
MER avg.	L/L_{vr}/d	3.6	2.9	3.4	2.5
Working Volume	L	9.5	6	6	6

Note: MER – Methane evolution rate

With reference to figure 3.5 the methane composition of the produced gas may be noted as the red highlighted zone. The lowest values are, as expected, in Reactor 1; the values increase in Reactor 2 and reach the highest values in Reactor 3. The final output gas will have a methane composition of c. 97 % which should comply closely with renewable methane standards. The process is deemed to work effectively at laboratory demonstration scale, which would be categorised as at Technology Readiness Level (TRL) 4; "technology validated in lab".
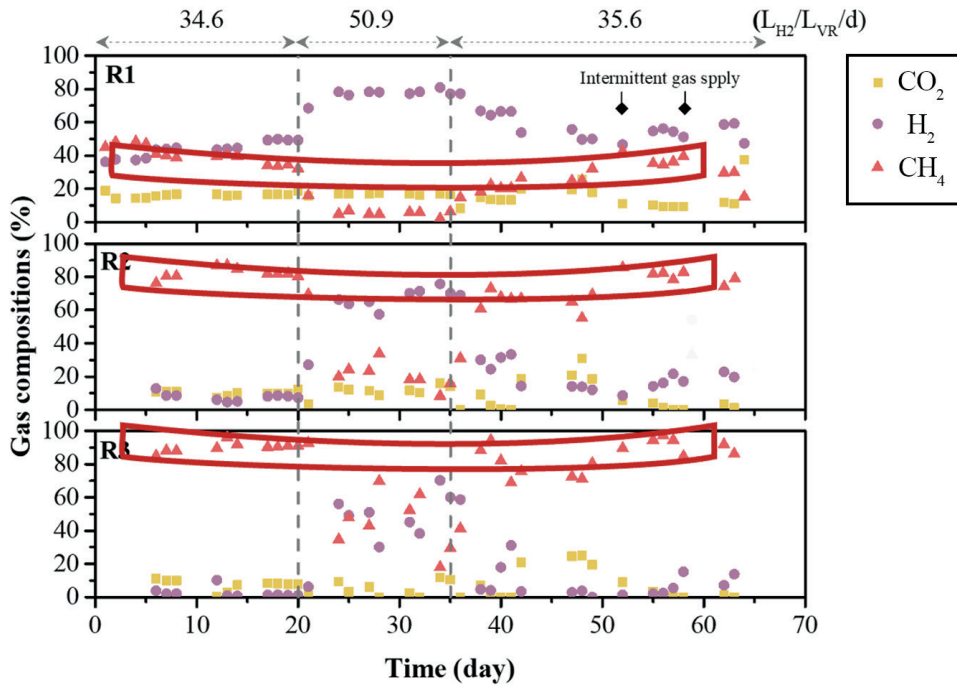

Figure 3.5: Composition of output gases output from continuous ex-situ biological methanation system

3.6 CHAPTER SYNTHESIS

Hydrogenotrophic methanogenic archaea are very good at converting H_2 and CO_2 to CH_4. The engineering technology to contain this biological process is available. There are limitations (that may be overcome) in ensuring that hydrogen dissolves into solution and in maximising the energy return per unit reactor size. These may be overcome by higher pressure systems (which may not always be facilitated in laboratory settings due to health and safety concerns) and fine dispersion systems. The next chapter will examine case stories of demonstration facilities at higher technology readiness level and systems that are close to commercialisation (TRL 8).

4 Applications of circular economy approaches to integration of anaerobic digestion with power to gas technologies

This chapter outlines a number of case stories which detail circular economy applications of power to methane in commercial settings, or at the very least demonstration scale. These include for catalytic and biological methanation processes. The last section describes a theoretical application which could be applied at any wastewater treatment facility of scale that requires oxygen.

4.1 AUDI E-GAS POWER TO METHANE FACILITY, WERLTE, GERMANY

The authors visited the Audi e-gas power to methane facility at Werlte, Germany in 2016 (Figure 4.1). This facility has been in operation since Audi opened the facility in June 2013. It is situated adjacent to a food waste digester which produces biomethane for gas grid injection. As such there is a concentrated, and continuous, stream of biogenic CO_2 available for use.

Three number 2 MW alkaline electrolysis cells (AEC) split water into hydrogen and oxygen through electrolysis, ideally using electricity from surplus offshore wind, from the North Sea. There is a hydrogen service station for hydrogen fuel cell cars, but this was not at the time in widespread use, due to a lack of hydrogen infrastructure. As such a major innovation here was the catalytic methanation stage which reacts the biogenic CO_2 (from the food waste digester) with the hydrogen from the electrolysers. The synthetic methane (termed e-gas) produced is identical to natural gas and as such may be used in compressed natural gas vehicles. The e-gas is injected into the gas grid in a similar fashion to the biomethane from the food waste digester.

The plant produces about 1,000 tonnes of methane per year or 1,400,000 m³ CH_4 (CH_4 has a density of 0.714 kg/m³) or 14 TWh/a of energy (CH_4 has a volumetric energy density of approximately 10 kWh/m³). The gaseous fuel is primarily destined for natural gas vehicles. If we assume an efficiency of 7 m³ CH_4/100 km per car, then this is sufficient for 1,000 cars to travel 20,000 km per annum.

Figure 4.1: Audi E-gas power to methane facility at Werlte, Germany; From clockwise top left: food waste digester; 6 MW alkaline electrolyser; hydrogen service station; overarching view of catalytic power to methane system

4.2 ALLENDORF, GERMANY

The authors visited this facility in September 2016; it is one of the first facilities which demonstrated power to methane and biological methanation technology at the same facility. The plant employs a PEM-electrolyser (Carbotech) and a biological methanation unit (BiON technology, MicrobEnergy GmbH). The methanation unit is a continuously stirred tank reactor with a volume of $5\,m^3$ which can operate at pressures between 5 to 15 bar and at temperatures between 50 to 80 °C (Figure 4.2).

The process was designed to treat $30\,m^3$ per hour of raw biogas; an assumption was made that this biogas is 53 % methane ($16\,m^3/h$) and 47 % CO_2 ($14\,m^3/h$). A 300 kW electrolyser with an efficiency of 60 % will generate 180 kWh of hydrogen or $60\,m^3$ of hydrogen per hour which is a little over 4 times that of the CO_2 from the biogas facility. With reference to Table 4.1 it may be noted that the output from the biomethanation facility increases the hourly methane output by 88 % through conversion of the CO_2 in the biogas to methane. We are informed that the resulting gas quality is greater than 98 % methane with less than 1.5 % H_2, with a feed in rate of methane of 400,000 kWh ($40,000\,m^3$ per annum) per year into the gas grid (IEA Bioenergy, 2018).

1. Proton Exchange Membrane electrolyser. 2. Methanation reactor 3. Measuring and mechanical process 4. Process and control

Figure 4.2. (a) Aerial view of power to methane facility at Allendorf, Germany (IEA Bioenergy, 2019) with (b) close up of biomethanation reactor (© Viessmann Werke GmbH & Co.KG)

Table 4.1: Estimate of input and output of power to methane facility at Allendorf

	Proton Exchange Membrane Electrolyser	Methanation reactor
Theory	$2H_2O = 2H_2 + O_2$ H_2 LHV of 3 kWh/m^3 or 33.33 kWh/kg	$4H_2 + CO_2 = CH_4 + 2H_2O$ CH_4 LHV of 10 kWh/m^3
Input	1 Litre of water per m^3 H_2 (Standard Temperature & Pressure (STP)) 300 kW electrolyser	$30\,m^3$ raw biogas / h Approximately $16\,m^3$ CH_4 & $14\,m^3$ CO_2/h
Output	300 kW at 60 % capacity = 180 kWh H_2/h = 60 m^3 H_2/h	$16\,m^3$ CH_4/h from biogas plus $14\,m^3$ CH_4/h from biological methanation facility

4.3 ELECTROCHAEA, DENMARK

The Electrochaea-BioCat demonstrator began operation at a wastewater treatment plant in Avedøre, Denmark in April 2016; it operated in excess of 4500 hours, until October 2019. This demonstration plant included a 1 MW electrolyser (Hydrogenics' S1000 alkaline electrolyser) integrated with a biological methanation facility (operating at 10 bar and 62 °C) with CO_2 sourced from a local biogas plant and included the use of raw biogas as well as purified CO_2. The design point of the plant is the utilisation of biogas at a rate of 125 m³/hour (75 m³/h CH_4 & 50 m³/h CO_2).

Electrochaea demonstrated the process in two configurations:

- As per Figure 2.2C they added pure streams of CO_2 post upgrading of biogas to the hydrogen to produce methane.
- As per Figure 2.2B they applied the technology as a method of upgrading biogas, converting the CO_2 within the biogas stream to methane. This, as mentioned previously, is more challenging due to the effect of methane in the biogas influent on the partial pressure of the hydrogen and carbon dioxide gases. It also requires a higher throughput of biogas, as 60 % of the biogas (CH_4) is not involved in the process.

Operation under both configurations was shown to offer CO_2 conversion efficiencies of more than 97 % and both configurations produced a similar gas output, which was injected into the local gas distribution grid (Liebetrau et al., 2020). With reference to Table 4.2 a stoichiometric ratio of 4:1 (H_2:CO_2) led to a design with a 1MW electrolyser producing 200 m³/h of hydrogen to react with 50 m³/h of CO_2 to produce 50 m³ of CH_4. The methane end product increased in yield by 67 %.

Implementing a circular bioeconomy approach, the heat from the exothermic reaction, both from biomethanation and from the electrolyser, was recycled to be used in the wastewater treatment plant.

Electrochaea facilities such as in Avedøre, Denmark and in Switzerland have demonstrated flexibility with load factor tests demonstrating operation between 0 and 100 % capacity. These facilities have shown rapid recovery after periods of shutdown; such intermittent operation is of great benefit in matching operation of an electrolyser with electricity sourced from variable renewable electricity. Electrochaea sees development of facilities in scale as per Table 4.3.

Figure 4.3. Photograph of BioCat Plant on BIOFOS site in Avedøre (from Power-to-Gas via Biological Catalysis (P2G-Biocat))

Table 4.2: Estimate of input and output of Electrochaea-BioCat power to methane facility at Avedøre, Denmark

	Alkaline Electrolysis Cell Electrolyser	Methanation reactor
Theory	$2H_2O = 2H_2 + O_2$ H_2 LHV of 3 kWh/m³ or 33.33 kWh/kg	$4H_2 + CO_2 = CH_4 + 2H_2O$
Input	1 MW electrolyser using 1000 kWh of electricity per hour	125 m³ raw biogas/h 75 m³ CH_4/h & 50 m³ CO_2/h
Output	1000 kW$_e$h at 60 % capacity = 600 kWh = 200 m³ H_2 /h	75 m³ CH_4 /h from biogas plus 50 m³ CH_4 /h from biological methanation facility

Table 4.3: Future scale of Electrochaea's BioCat biological methanation facilities (from Electrochaea GmbH data sheets)

	BioCat 1	BioCat 10	BioCat 50
Electrolyser	$1MW_e$	$10\ MW_e$	$50\ MW_e$
Reactor Temperature and Pressure of 63 °C at 10 bar with CO_2 conversion efficiency of greater than 97%			
Gas Input	$200\,m^3/h\ H_2$ $50\,m^3/h\ CO_2$	$2000\,m^3/h\ H_2$ $500\,m^3/h\ CO_2$	$10{,}000\,m^3/h\ H_2$ $2500\,m^3/h\ CO_2$
Installed power at methanation	45 kW	370 kW	1600 kW
Outputs Grid quality gas Thermal energy Metabolic water	$50\,m^3/h\ CH_4$ $130\ kW_{th}$ 80 L/h	$500\,m^3/h\ CH_4$ $1275\ kW_{th}$ 800 L/h	$2500\,m^3/h\ CH_4$ $6400\ kW_{th}$ 4000 L/h
Footprint	$150\,m^2$	$480\,m^2$	$1070\,m^2$

4.4 OPERATION OF A CIRCULAR ECONOMY, ENERGY, ENVIRONMENTAL SYSTEM AT A WASTEWATER TREATMENT PLANT

A major consideration in all developments of waste treatment facilities and new infrastructure is public acceptability. There are significant targets for hydrogen production into the future. For example, Ireland (population of 5 million people) has a 2030 target of 2 GW of hydrogen produced from offshore wind. Where do we build these electrolysers? How do we minimise potential for planning objections and public disquiet? How do we maximise the function of the electrolyser, including for valorisation of oxygen? We should not develop an electrolyser purely for the sake of developing an electrolyser and producing hydrogen. We should optimise the role of the electrolyser within a circular economy, energy, and environment system.

There can be significant benefits in positioning an electrolyser within an existing wastewater treatment facility as opposed to on a green field site. Wastewater treatment facilities tend not to be tourist attractions or even allow access to the public; they are generally situated away from public or leisure amenities. Many of the processes include for wet organic materials such as wastewater and sludges, which tend to be associated with bad odours; such facilities include for significant mechanical and electrical facilities and many wastewater treatment facilities of scale already have anaerobic digestion installed for sludge treatment. They tend to be well connected to utilities such as water (required for electrolysis), the electricity grid (required for electrolysis) and as biomethane is produced it would be beneficial to be near the gas grid (to inject produced synthetic methane or biomethane). In terms of planning authorisation, it could not be said that there is a significant change of use and in terms of available utilities wastewater treatment facilities could be said to be an optimal site for new electrolysers.

The electrolyser produces oxygen that may be used in the aeration basin instead of air. This can reduce the pumping requirements at a wastewater treatment facility by a factor of 5 (oxygen comprises 21 % of air). Existing sludge digesters produce biogas which have some uses such as heating digesters and production of electricity. However, the hydrogen produced in the electrolyser may be used in an ex-situ biological methanation process to react with the CO_2 in the biogas and generate pure biomethane, whilst increasing the methane yield by 50 to 60 %. Biomethanation is an exothermic process and as such the heat (along with heat generated by the electrolyser) may be used to heat the digesters replacing the previous biogas function.

Rusmanis et al. (2022) proposed a system as illustrated in Figure 4.4. The starting point was to position the electrolyser at the existing wastewater treatment facility for Cork City, Ireland which has a population equivalence (PE) of about 430,000. It was decided that the overarching design principle employed would be to satisfy the oxygen demand of the aeration basin (approximately 7,000,000 m^3 O_2 per annum). Pumping oxygen in the aeration basin rather than air could reduce the energy footprint of the facility by

40 %. This oxygen would be generated by a 10 MW electrolyser which operating at an 80 % capacity factor would require 70 GWh/a of electricity. Typically, renewable electricity from wind turbines is dispatched down due to curtailment (temporal imbalances between supply and demand) and/or constraint (due to inadequacy of the existing electrical infrastructure to transport the produced electricity). The electricity required for the 10 MW electrolyser would equate to the expected dispatch down of a 144 MW wind farm and as such the electrolyser can facilitate such a wind farm in maximising its electricity output and minimising its dispatch down. The 10 MW electrolyser would produce 14,000,000 m³ of hydrogen. The methane element of the existing biogas produced from sludge digestion could be increased by 55 % by the operation of an ex-situ biological methanation process which would use 22 % of the H_2 produced by the electrolyser. The biomethane produced could fuel 90 compressed natural gas trucks; the hydrogen produced (not used in ex-situ biomethanation) could fuel 300 fuel cell trucks. Obviously the hydrogen produced could serve other functions as arise in bespoke geographic specific communities.
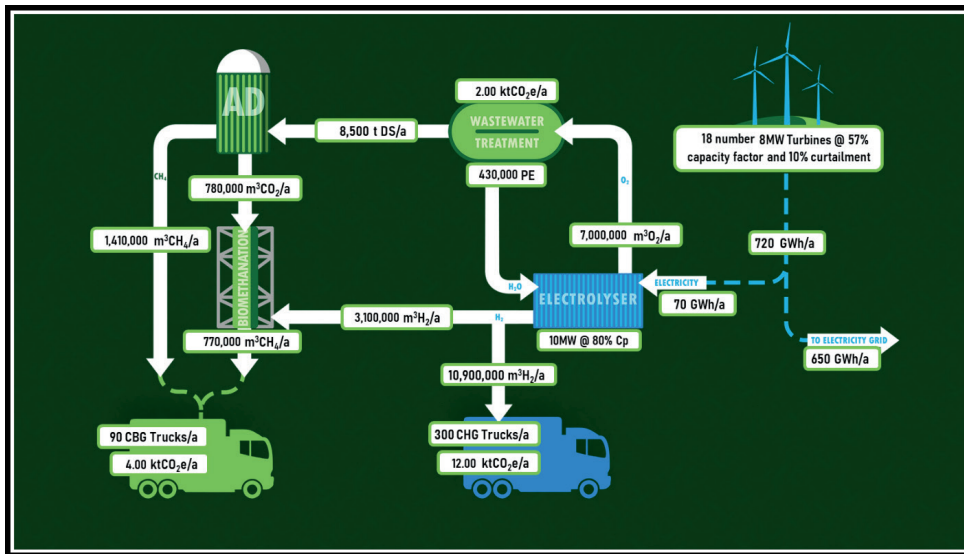

Figure 4.4. Circular economy, energy, and environmental system at a wastewater treatment plant, adapted from Rusmanis et al., (2022)

4.5 CHAPTER SYNTHESIS

There are applications of power to methane in place, the science is well understood and the process, as defined by the conversion efficiency, is close to optimisation. It may however be said that as of now, it is not a technology that is very well known or widely employed in situations where it would have merit. There are commercial pioneers of the technology in the market, and the vision for applications in the future has been described. In the authors' opinion such applications of this technology will follow with the advent of the new hydrogen industry. As electrolysers become widespread with the advent of the hydrogen industry, then biomethanation will follow suit. An application that in the authors' perspective would be hugely beneficial, is use of biomethane in the production of steam (a source of process heat) for the food and beverage industry, followed by the capture of CO_2 from the exhaust stack, and combination with hydrogen to produce low carbon energy vectors such as power to methane or indeed, power to methanol as discussed in the next chapter.

5 Further Power to X technologies

This chapter will introduce other Power to X technologies. The depth of analysis employed in explaining biomethanation in the previous two chapters will not be used here but rather a perspective will be presented as to how these technologies will benefit from interaction with anaerobic digestion and to assess what the technologies may look like in the coming decades.

5.1 POWER TO METHANOL

Methanol is the second most produced chemical on the planet with a worldwide production of 110 million tonnes per annum. At present it is used in the production of acetic acid, formaldehyde, acrylic plastics, fabrics and fibres for clothing, adhesives, paints, and chemical agents in pharmaceuticals[8]. In a future decarbonised world, it is expected to be a major source of shipping fuel; this will lead to increased worldwide production. Methanol (CH_3OH) is typically produced from natural gas; this has been problematic of late (2022/2023) due to the scarcity and price of natural gas as a result of the conflict in Ukraine and sanctions against Russia. Beyond security of supply issues, a net zero world will necessitate all existing functions of methanol and future demands on methanol to be renewable with as low a carbon footprint as possible. It is suggested that this will place significant demands on the resource of hydrogen from renewable electricity and on the resource of biogenic CO_2.

The similarity of the chemical representation of methanol (CH_3OH) and methane (CH_4) highlights the potential for biogas and biomethane to be used in the production of methanol. With reference to Equation 5.1 we can see how biogas with 75 % CH_4 and 25 % CO_2 may be converted to methanol. Of issue is that this is not the typical composition of biogas. This may be overcome by splitting biogas into carbon monoxide (CO) and H_2 using an electrically driven catalytic convertor and then to react CO with renewable hydrogen. Methanol synthesis as per equation 5.2 is an exothermic reaction conducted between 200 and 300 °C and 3.5 to 10 MPa (IEA Bioenergy, 2020b).

$$3CH_4 + CO_2 + 2H_2O \quad = \quad 4CH_3OH \qquad\qquad \text{Eq. 5.1}$$
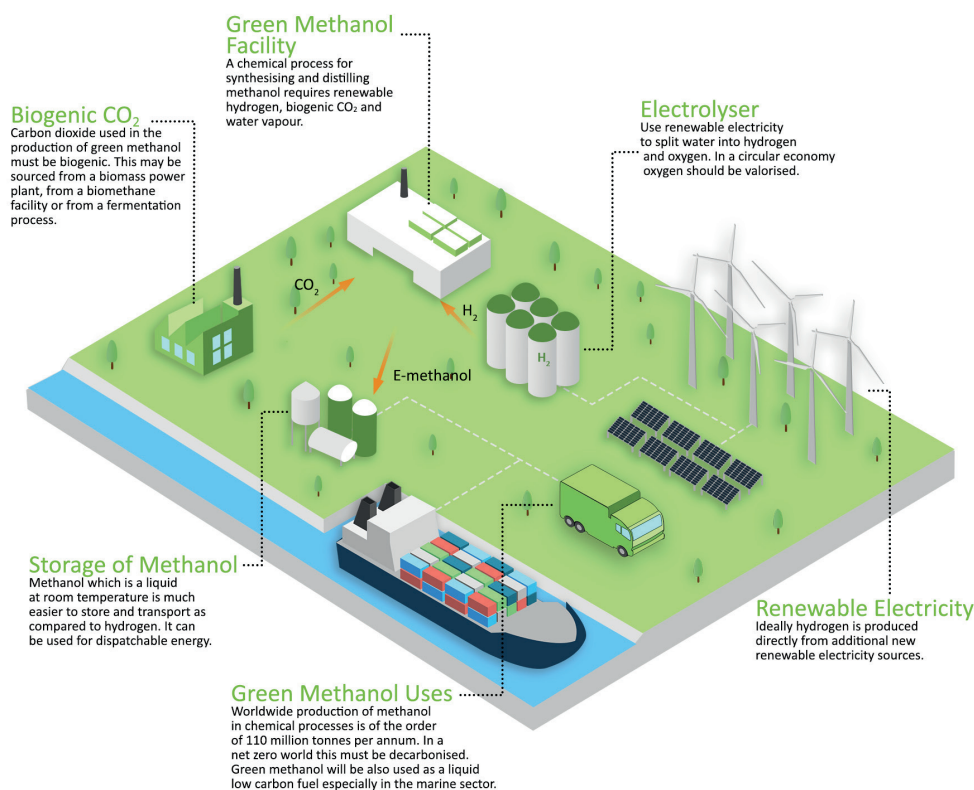$$CO + 2H_2 \quad = \quad CH_3OH \qquad\qquad \text{Eq. 5.2}$$
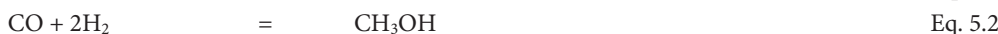
The Danish logistics firm Maersk has 19 methanol ready dual fuel ships set to be put into operation in the next three years. Of issue at the moment (2023) is the lack of green hydrogen derived methanol on the market. As a transition Maersk have signed a deal with OCI (a Danish green fuel and fertiliser producer) to supply a low carbon methanol for the maiden voyage of its first ship from Korea to Copenhagen. This low carbon methanol will be synthesised from biomethane[9]. The carbon will be biogenic but there is a scarcity of green hydrogen. A decarbonised shipping industry fuelled by green methanol will need significant quantities of green hydrogen generated from water electrolysis using renewable electricity. Of further issue with shipping is the logistics and distribution of green fuels. The requirement for bunkering such renewable fuels at strategic ports across the planet to facilitate long voyages is crucial. It complicates the prospects of a short term green methanol industry as it is not one port that must make the decision and investment, but all major ports. It may be that biogenic carbon and renewable hydrogen will need to be produced at the major shipping ports, rather than export of biomethanol from a few strategic developments.

In terms of future developments, "Orsted" are in the process of developing a 50,000 tonne per annum green methanol facility in Sweden using a 70 MW electrolyser, and Iberdola (Figure 5.1) have plans for a 300,000 tonne per annum green methanol facility in Australia[8].

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[8] https://www.mgc.co.jp/eng/rd/technology/methanol.html#:~:text=Methanol%20and%20its%20derivative%20 products,agent%20in%20pharmaceuticals%20and%20agrichemicals.
[9] https://www.hydrogeninsight.com/transport/maersk-will-use-green-methanol-for-dual-fuel-ships-maiden-voyage-but-it-wont-be-made-from-green-hydrogen/2-1-1466159

Figure 5.1. Green Methanol production system, adapted from:
https://www.iberdrola.com/documents/20125/2056775/metanol-verde-infografia-EN.pdf

5.2 POWER TO AMMONIA

Worldwide production of ammonia (NH_3) is of the order of 160 million tonnes; this is responsible for the release of 450 million tonnes of CO_2 through ammonia synthesis which corresponds to about 1 % of global CO_2 emissions (Boerner, 2019). Ammonia is the most produced chemical on this planet; it is the source of nitrogen fertiliser. The source of the hydrogen molecules in present day production of ammonia (NH_3) is fossil fuel (such as from the hydrogen molecules in natural gas (CH_4)) so as such we spread fossil fuels on our agricultural land to increase the yields of food to feed our ever-growing population. If the intention is to decarbonise agriculture in a net zero world, we must employ low carbon fertilisers and we must produce green ammonia. Even if the actual fertiliser were produced in a low carbon manner, there is still the challenge of dealing with nitrous oxide (N_2O) release from soil in the process of fertilising agricultural land. N_2O has a global warming potential of 265 times that of CO_2; net zero will not be easy. Planet Earth has about 0.2 ha of arable land per person. Arable land is finite and as the population grows, that land must yield more food per unit area for more people. The demand for fertiliser may not reduce. Beyond fertiliser, ammonia is used for textiles, plastics, explosives, and in the pharmaceutical industry. Considering ammonia is also mooted as a decarbonised fuel and/or a means of transporting hydrogen across the planet, then we can understand the massive demand for renewable hydrogen molecules.

The Haber Bosch process reaction as synthesised in Equation 5.3 runs at temperatures of about 500 °C and pressures of 20 MPa (over an iron catalyst); the reaction is slightly exothermic.

$$N_2 + 3H_2 \quad = \quad 2NH_3 \quad\quad\quad\quad\quad\quad\quad\quad\quad\quad\quad\quad\quad\quad\quad\quad\quad\quad \text{Eq. 5.3}$$

For green ammonia the hydrogen molecules must be green. There is potential for biomethane to replace natural gas in the generation of hydrogen molecules for the Habor Bosch process. However, the demand for fertiliser on a worldwide scale may be too high for biomethane to play a significant role. It is more likely that hydrogen will be sourced from electrolysers powered by renewable electricity. This then

becomes a further challenge in deciding decarbonisation pathways. What hydrogen pathway gets priority? Is it better to use hydrogen for methane production, for methanol production or for ammonia production? A major benefit in terms of the resource of power to ammonia is that the process is not constrained by the availability of biogenic CO_2 but rather can use nitrogen from the atmosphere.

A green ammonia demonstration facility including for collaboration between Siemens Energy, UK, Oxford University and Cardiff University uses the Haber-Bosch process with hydrogen sourced from wind derived renewable electricity (Figure 5.2). For further developments in green ammonia and innovative processes to reduce the carbon footprint of green ammonia the reader is referred to Boerner (2019).

Figure 5.2. Green Ammonia Production System, from (Boerner, 2019) Credit: Siemens Energy

5.3 CHAPTER SYNTHESIS

There are viable Power to X options in producing hydrogen, methane, methanol, and ammonia, but as yet there are no definitive agreed pathways. The final chapter explores where Power to X technologies should or could play a role in electricity, heat, and transport sectors.

6 Discussion and Conclusions

6.1 ROLE OF ELECTRICITY AND POWER TO X IN ELECTRICITY, HEAT AND TRANSPORT

There are many options and pathways to decarbonisation. The solutions can be bespoke and may depend on policy or geopolitical considerations. There is a significant support for electrification of energy where possible and decarbonisation of electricity may then lead to decarbonisation of energy. Examples include battery electric vehicles for private transport and heat pumps for heating and cooling. In the last number of years, hydrogen has received a very positive press, but there is still discussion as to what hydrogen is best suited for when it comes to end use applications.

There have been proposals for hydrogen to be utilised as a source of heat (such as in hydrogen boilers for industry) but the route from renewable electricity to hydrogen via electrolysis, including for compression, storage and distribution is expensive. Numerous sources of literature are available on future levelized costs of energy (LCOE) of hydrogen and in the short term pre-2030, analysis would suggest costs of € 3.50/kg (or 10.5 c/kWh) are optimistic (McDonagh et al. 2020). Contrast this complex expensive process with combustion of wood chips which would probably be available for less than 5 c/kWh. The authors suggest that hydrogen as a source of renewable heat is not always economically prudent, but cost may only be one consideration in bespoke solutions for sustainable energy provision in a net zero world.

There have been proposals for hydrogen as a means of energy storage when electricity is produced at periods of low demand for electricity and as such the electricity would have been otherwise dispatched down; in the hydrogen economy this "surplus" electricity may now be converted to hydrogen via electrolysis. Of issue here is expense and efficiency. McDonagh et al., (2020) considered a hybrid system where offshore wind could produce electricity and hydrogen. The model is such that when electricity demand is significant, then electricity is sold, but at times of low electricity demand (times when electricity supply is equivalent to demand and including times where supply is more than demand where curtailment would take place) hydrogen would be produced. As such, cheaper electricity is used to produce hydrogen. The electrolyser will have an optimistic efficiency of 70 %, so for example electricity at 4.5 c/kWh (which would be considered cheap) will incur an electricity cost to the hydrogen of a minimum of 6.5 c/kWh. However, when we consider electrolysers, storage and distribution, electricity at 4.5 c/kWh is modelled as producing hydrogen at a cost of about $ 3.50/kg or 10.5 c/kWh (McDonagh et al., 2020), allowing for downtime of the electrolyser.

To convert back to electricity, we may consider a fuel cell system operating at, optimistically, 70 % efficiency. As such the round cycle efficiency would be of the order of 49 % (70 % * 70 %) meaning the electricity cost element of the hydrogen would be double the price of the electricity purchased. If we consider hydrogen produced from electricity at 4.5 c/kWh costing 10.5 c/kWh of hydrogen, then the very minimum price of electricity produced from hydrogen would be 15 c/kWh (10.5 c/kWh / 0.7); this simple analysis ignores costs of fuel cells and associated electrical and hydrogen distribution. We can readily say that electricity from hydrogen will cost about 3 to 4 times the cost of the electricity it is produced from. There are however bespoke geopolitical reasons that systems may have applications despite disadvantages such as cost and efficiency. We do need to look at the entire energy system over time. In a decarbonised world free of fossil fuels, we will need dispatchable electricity. As an example, Ireland in the winters of 2009 – 10 and 2010 – 2011 experienced prolonged cold spells when wind generation of electricity was very low in contrast to the electricity demand (Craden et al., 2018). In these periods, in a future decarbonised world, with little dispatchable electricity, hydrogen storage of about 20 days would be extremely beneficial for dispatchable power generation even though the electricity would be very expensive; the alternative would be blackouts. The entire energy system could support the expense of such storage and use of hydrogen to support the electricity grid.

When we consider transport there is a very strong case for electrification of light vehicles. Previous research suggests, that for 100 kWh of electricity, a battery electric vehicle will transfer 70 kWh to propulsion, while for a hydrogen fuel cell vehicle this is limited to 22 kWh. Gray et al., (2022) would suggest that

batteries are preferable to hydrogen fuel cells for haulage with ranges below 450 km. Beyond 450 km, the weight and volume of the battery are too great and impact on the carrying capacity of heavy goods vehicles. Even though the electricity demand to power the truck increases by a factor of three (Figure 6.1), the hydrogen fuel cell is preferred.

What is the role of power to methane in transport? The energy conversion efficiency of power to methane has two overarching elements. Firstly, the efficiency of electricity conversion to hydrogen which is expected to be of the order of 70 %. The next step is conversion of hydrogen to methane. With reference to the Sabatier equation ($CO_2 + 4H_2 \rightarrow CH_4 + 2H_2O$) it may be noted that 4 m³ of H_2 (4 * 3 kWh/m³) is required to produce 1 m³ of CH_4 (10 kWh/m³) and as such the maximum conversion efficiency possible would be 83 % (10 kWh/m³ CH_4 / 12 kWh/m³ H_2), while if we include for methanation parasitic losses, the conversion could be reduced to of the order of 80 % (system specific). Thus, the conversion from electricity to methane could have efficiencies between (0.7 * 0.8 =) 56 % and (0.7 * 0.83 =) 58 %. In powering heavy goods vehicles Gray et al., (2022) suggest the electricity required to fuel 1 vehicle on synthetic methane produced via the power to methane pathway was nearly 5 times that of the battery electric vehicle and almost twice that of the hydrogen fuel cell electric vehicle (Figure 6.1). This takes into consideration the lower efficiency of an internal combustion engine (ICE) as compared to a Fuel Cell Vehicle (FCV). There is potential for the methane to be used in a fuel cell, which could reduce this differentiation in electrical demand somewhat, but not anywhere approaching parity.
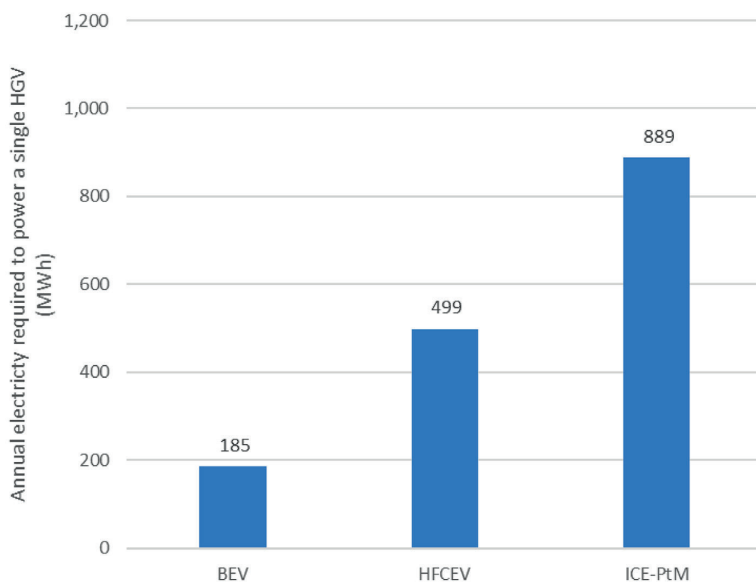
Figure 6.1. Electricity supply required to power a Heavy Goods Vehicle (HGV) as a battery electric vehicle (BEV), a hydrogen fuel cell electric vehicle (HFCEV), an internal combustion engine power to methane (ICE-PtM) from (Gray et al., 2022)

In examining long distance shipping, neither batteries nor hydrogen are deemed appropriate as volume and weight impact significantly on carrying capacity (Gray et al., 2021). Ships will need renewable liquid fuels; this brings us back to methanol and ammonia. The perspective seems to suggest that methanol is preferable as ammonia has issues with toxicity which in the event of an accident may be deleterious to the health of the sailors.

Therefore, it may be asked where the ideal application of power to methane as a transport fuel is? It does not compete on a theoretical energy analysis with a battery electric vehicle; nor does it compete with a hydrogen fuel cell truck. For large ships and planes, liquid fuels are required or preferred. However, all solutions are bespoke and as such we need to assess the total cost of ownership of a fuel system and the bespoke problem set. If we wish to have battery electric heavy trucks travelling 450 km across our countries,

we will need significant electrical infrastructure. In assessing new renewable fuels, we need to consider the total cost of ownership including for infrastructure, the cost of the vehicles, the availability of the vehicles, the fuelling or charging infrastructure for the vehicles, and the ability to roll out such systems in a short time period. This is not always easy; for example, in Dublin, Ireland, at present (2023) 130 electric buses (purchased in 2022) lie idle in bus depots as the charging infrastructure is not in place[10].

6.2 ALL SOLUTIONS ARE LOCAL

Just as all politics are local it may be said all solutions are local. We don't always do what is most efficient. If for example, we wished to be sustainable in transport, we would walk and cycle and only drive where absolutely necessary. But we drive. If it is a given that we will continue to drive and we want to be most efficient, then we would drive small electric cars, but sales in SUVs are increasing as a portion of all car sales. As such, it may well be that biomethane is chosen as an optimal practical solution for a particular application. In Linkoping, Sweden, the bus service operates on biomethane. There is a significant history dating back to the last century of using biomethane in transport both in buses and in cars. The source of the biomethane is residues, such as food waste, slaughter waste, and municipal sludges from wastewater treatment. As such a circular economy, energy, and environment system is employed with numerous local benefits, including biofertiliser production, clean air, and local employment. Over the last few years, a liquified biomethane system has been used to fuel trucks distances of up to 1000 km. In Linkoping, the buses and trucks fuelled by biomethane are Swedish while the electric buses recently introduced to Linkoping are produced abroad. Experience in Sweden has shown that electric buses need to import heating in the buses, which is an extra fuel system; this is not required in a biomethane fuelled bus. Details on biomethane as a transport fuel within a circular economy, energy, and environmental system are detailed in Ammenberg et al., (2021). If the infrastructure is in place, it is relatively straight forward to expand on such a system. This would seem to be an ideal application for power to methane. Addition of an electrolyser and ex-situ biomethane system to the biogas facility could add 60 % to the renewable fuel produced, and may well be the optimal solution to a bespoke problem set. The costs of biomethane will be lower than that of power to methane as approximately 60 % of the methane will be associated with biological conversion of residues to biomethane, as opposed to conversion of hydrogen to methane.

Another bespoke example is in Denmark where the gas grid is presently (2023) at 39 % renewable green biomethane gas. The addition of electrolysers and biomethanation systems at these digesters could add 60% to the renewable gas resource and bring the renewable supply to 62 % without adding any more feedstocks. Denmark has a target of 100 % green gas by 2035 including for such power to methane applications (IEA Bioenergy, 2019b).

REPowerEU (2022) has set a target for biomethane of 35 billion cubic meters of biomethane by 2030. This is a 10-fold increase on present (2023) levels of production. There are concerns about the level of sustainable feedstocks in reaching this target. Utilising power to methane systems (integrated anaerobic digestion with Power to X technologies) could reduce this required feedstock resource by 40 % through generation of a standard model of biogas plant, electrolyser, and ex-situ biomethane facility.

Power to X is a series of technologies that help in the transition to a decarbonised world. We will not electrify all energy sources and if we could electrify all energy, we would still have emissions from agriculture and waste management. The ambition of this report is to ensure that technology developers, undergraduate students, utilities, and policy makers are aware of the role that Power to X can play. To optimise pathways to net zero, it is imperative that policy makers, municipalities and utility providers align their regional plans for decarbonisation with the present state of technology, to ensure proactive implementation of Power to X systems and to facilitate maximisation of circularity of energy and environmental systems.

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[10] https://www.independent.ie/regionals/dublin/dublin-news/why-more-than-130-new-electric-buses-bought-a-year-ago-are-still-lying-idle-in-dublin/a894095440.html

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