Integration of Power to Methane in a Waste Water Treatment

Plant – A Feasibility Study

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Abstract

The integration of a biomethanation system within a wastewater treatment plant for conversion of CO₂ and H₂ to CH₄ has been studied. Results indicate that the CO₂ could be utilised to produce an additional 13,420 m³ / day of CH₄, equivalent to approximately 133,826 kWh of energy. The whole conversion process including electrolysis was found to have an energetic efficiency of 66.2 %. The currently unoptimised biomethanation element of the process had a parasitic load of 19.9 % of produced energy and strategies to reduce this to <5 % are identified. The system could provide strategic benefits such as integrated management of electricity and gas networks, energy storage and maximising the deployment and efficiency of renewable energy assets. However, no policy or financial frameworks exist to attribute value to these increasingly important functions.

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1. Introduction

Society faces a number of challenges relating to future energy production and transmission as efforts continue to reduce global CO₂ emissions. Whilst deployment of renewable electricity generation technologies has increased significantly in the UK from 6.8 % of total generation capacity in 2010 to 24.6 % in 2015 (BEIS, 2016), grid constraints (Van den Bergh et al., 2015) and high integration costs (Hirth et al., 2015) may limit the effectiveness of power grids based on high penetration of renewables. These problems are not unique to the UK but are repeated throughout developed countries with a high penetration of renewable electricity production and reduction in fossil energy production. Power to Gas (PtG) is an approach that, if successfully developed and deployed, would allow the inter-operability of electricity and gas grids, maximise the productivity of renewable electricity generation infrastructure, contribute towards the decarbonisation and long term viability of regional or national gas transmission networks, and allow for energy storage within the gas grid. The integration of renewable energy generation, energy storage and waste management operations may also bring additional benefits such as the improved efficiency of waste management processes.

PtG is based around the electrolytic production of hydrogen, and, where this electrolysis is driven by renewable electricity, it offers a route for producing low carbon fuel gases (Jensen et al., 2007, Carmo et al., 2013). Whilst hydrogen may represent a valid fuel vector in the long term, at present in many countries it cannot be added to natural gas

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infrastructure in significant quantities – for example in the UK current gas quality legislation only permits <0.1 % hydrogen (UK Government, 1996). The potential for including hydrogen concentrations of 0.5-20 % are being considered (HSE, 2015), however, these higher levels may be limited only to some networks, and utilising hydrogen at 20 % by volume would deliver only 6 % of the energy of the same volume of methane. Conversion to high hydrogen percentages is likely to be focused on urban populations and will require significant changes to the gas network infrastructure and end use appliances (Leeds City Gate, 2016). Approaches that utilise low carbon hydrogen to produce low carbon synthetic methane, which is fully compatible with current gas grid infrastructures and regulations, therefore have the potential for short to medium term deployment i.e. over the next few decades, and could contribute to long term viability of more spatially distributed networks serving lower population densities. The Sabatier process thermo-chemically reacts H2 with CO2 to produce CH4 (Jürgensen et al., 2015), however the process operates at high temperatures (>250 °C) and pressures (>10 bar) and can be difficult to control, utilises expensive metal catalysts, and is not compatible with the intermittency of renewable energy supplies. Alternatively, several microbial species (hydrogenotrophic methanogens) have the ability to utilise hydrogen in combination with carbon dioxide to produce methane and water e.g. (Savvas et al., 2017a), as summarised in the equation (1).

Equation (1): $CO_2 + 4H_2 \rightarrow CH_4 + 2H_2O.$

Another two stage anaerobic microbial pathway could also catalyse this conversion through a combination of homoacetogenesis and acetotrophic methanogenesis (Savvas et al, 2017b).

Whilst the biomethanation process is still at early stage research and development, the approach offers a means of potentially achieving high conversion efficiencies at low temperatures and pressures and is tolerant of contaminants typically found in biologically produced gases or flue gases.

Previous research has conceptualised several applications and configurations for such a biological process, including the production of high methane content biogas from anaerobic digestion plants (Luo et al., 2012, Bensmann et al., 2014), the operation of standalone hydrogenotrophic reactors (Luo and Angelidaki, 2012, Bassani et al., 2015), using single strain microbial populations (Martin et al., 2013) and mixed culture approaches (Savvas et al., 2017a). More recently, some research has been undertaken to evaluate the potential for actually deploying such technology at industrial scale, and quantifying the environmental burdens / benefits that this might bring. Götz et al. (2016) undertook a techno-economic assessment of various Power to Methane (P2M) approaches and concluded that biological methanation, whilst easier to control than chemical catalytic approaches, was limited in effectiveness by poor H₂ mass transfer to the liquid phase. This was also a topic of discussion by Savvas et al, (2017b) and further improvements were achieved by the development to a novel biofilm plug-flow reactor capitalising on lower energy requirements of gas to gas (reduced liquid layer) transfers. By undertaking a regional mathematical modelling approach, Zoss et al. (2016) concluded that wind resources in the Baltic states would be insufficient to generate enough H₂ to utilise the CO₂ produced in the regions' biogas plants, but recognised the important grid balancing role that such an approach would make. In assessing the life cycle burdens of P2M and Power to Syngas compared to fossil fuel reference cases,

Sternberg and Bardow (2016) concluded that where availability of renewable electricity was limited, syngas production from fossil sources had lower environmental burdens than other options, highlighting the importance of renewable electricity in the viability of P2G systems. The importance of utilising a high proportion of renewable electricity to drive a Power to Gas process was also stated by Reiter and Lindorfer (2015) in their assessment of life cycle Global Warming Impacts of PtG options. Walker et al. (2016) concluded that PtG (in this case hydrogen) could be cost competitive with fossil reference processes providing that the low carbon nature of the hydrogen and the function of providing an energy storage mechanism is reflected in the pricing structure. Gutiérrez-Martín and Rodríguez-Antón (2016) evaluated the technical and economic feasibility of Power to Methane for energy storage based on a thermo-chemical catalytic methanation stage. Vo et al. (2017) assessed the feasibility of matching curtailed wind electricity in Ireland with CO₂ produced in biogas plants to biologically produce methane for transport fuel use, and concluded that predicted 2020 curtailed electricity would be sufficient to utilise 28.4% of CO₂ available in biogas plants.

1.1 Study Aims

This study aims to evaluate the feasibility of integrating a novel biomethanation system at a waste water treatment plant (WWTP) incorporating sludge digestion, biogas production, biogas upgrading and gas grid injection. This application was chosen as a potential early adopter of PtG / biomethanation technology as it has an abundant supply of CO₂ and is likely to have on site uses for process products including oxygen (in the aeration processes) and methane (in on site CHP or gas grid injection facilities), and the water industry is familiar with the operation of industrial biological processes. A

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number of water companies are also investing in the deployment of other renewable energy generation assets near WWTPs. Study conditions, industrial practice and regulatory frameworks are based on those found in the UK, but are broadly applicable in other developed countries. If integration is feasible, wastewater treatment plants could provide novel and increasingly strategic roles within society by delivering the following functions; (i) treatment of wastewater, (ii) recovery of nutrients, (iii) production of biogas, (iv) recycling of CO₂ (via biomethanation), (v) balancing / integration of power and gas grids (via biomethanation)

This is the first time that this approach to biomethanation has been evaluated for full scale deployment in an industrial waste water treatment plant.

The scope of this study covers the following points:

- To numerically scale up a laboratory based biomethanation process to investigate the configuration and integration of such a process into a waste water treatment / sludge digestion process.
- 2. To quantify the broad operating parameters of such a system based on current knowledge and experience, in particular the energetic requirements of the system. This is undertaken by considering current operating parameters of both the WWTP and the PtG / biomethanation system and by calculating the major material flows and energetic requirements.

2. Methods

2.1 System Boundary

The boundary of the system studied is shown in Figure 1. The energy requirement of the integrated process is evaluated by quantifying primary energy input to electrolysers (hydrogen production), the biomethanation process, the energy available through methane production, excess thermal energy and, where appropriate, energy savings from utilisation of co-products (i.e. oxygen). As CO₂ for the biomethanation process is sourced from biogas, the existing biogas production and upgrading facilities are included in the overall integration evaluation.

2.2 Primary Elements of WWTP

The WWTP considered in this study is consistent with a large conventional sewage treatment plant treating municipal wastewater generated by a population equivalent of approximately 900,000 and treating approximately 65.7 million m³ of sewage per year. The primary elements of the WWTP are shown in Figure 2. Of specific interest is the presence of secondary treatment methods that rely on the aeration of mixed liquors in both the activated sludge plant and sequencing batch reactors that currently utilise air as the oxygenating agent. The integration of PtG / biomethanation into such a process has the potential to supply pure oxygen that could be utilised in a modified aeration system, therefore potentially reducing operational energy consumption. In the context of this study, aeration of wastewater is assumed to be required to achieve a reduction in BOD from 220 mg/l to 20 mg/l, and a reduction in Nitrogen from 40 mg/l to 20 mg/l.

In addition, the presence of sludge digestion facilities and the generation of biogas provide a large source of biogenic CO₂. The volume of biogas produced from the digestion of sewage sludge is approximately 40,000 m³/d. Given an average methane

content of 62.5 %, this gives an approximate CO₂ production of 14,720 m³ / d. Off gas from the biogas upgrading plant (based on water scrubbing) typically comprises of approximately 90 % CO₂ and 8.2 % CH₄, with the remaining 1.8 % being made up of low concentrations of N₂, H₂, CO, H₂S and O₂ (Malmberg Water AB, 2015) and is therefore considered as a viable feed gas for biomethanation.

2.3 Electrolytic Hydrogen Production

Polymer Electrolyte Membrane (PEM) electrolysis was considered the most compatible with the system goals, primarily due to the ability to vary output according to power availability or demand, and the high quality of hydrogen produced. A 1030 kW PEM electrolyser was considered as one of the larger industrial scale, market available electrolysers. This was assumed to deliver a nominal hydrogen production of 462 kg (5,139 m3 NTP) of 99.99% hydrogen per 24 hours of operation with an input of 40 litres of water per kg of hydrogen produced and a hydrogen outlet pressure of 20 bar. For the purposes of this study it is assumed that electrolyser performance is maintained over the working life of the plant.

Total CO₂ production from the digestion of sewage sludge at the WWTP is 14,720 m³ / day. Based on achieving 98 % stoichiometric conversion within the biomethanation plant and an electrolyser technical availability of 90 %, a hydrogen supply of 4,574 kg / day (50,877 m³ / day NTP), which could be delivered by 11 No. 1030 kW electrolysers operating at nominal output, would be sufficient to utilise 14,350 m³ / d CO₂. Oxygen would also be produced at a rate of 8 kg O₂ / kg H₂ (0.54 m³ O₂ / 1 m³ H₂ NTP) and the study considered the feasibility of diverting this oxygen to the secondary treatment stage of the WWTP to replace a proportion of the air utilised as an oxygenating agent.

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2.4 Proposed Biomethanation Parameters

The biomethanation approach considered in this study was that developed at laboratory scale by Savvas et al. (2017a). The process (Fig. 3) comprises an upright column reactor containing a lithotrophic methanogenic enriched mixed culture initially inoculated with anaerobically digested sewage sludge, and was operated at mesophilic temperature (37 °C) and ambient pressure. This liquid medium was re-circulated using a centrifugal pump via a low level outlet in the reactor wall, and was re-introduced close to the top of the reactor. Pre mixed feed gases (H₂/CO₂) were introduced at a volumetric ratio of 78 % H₂ / 22 % CO₂ to the bottom of the reactor vessel. High mixing rates, and the associated breaking of gas bubbles by the centrifugal pump, were shown to increase gas/liquid transfer therefore maximising methane yields, and an optimum (for CH₄ yield) liquid recirculation rate of 6 No. reactor volumes per minute was operated at laboratory scale.

Reactors were operated at steady state for a number of feed gas rates. At laboratory scale and during the period of enrichment phase of the microbial culture the biomethanation process produced methane gas qualities at the outlet of 98.9 % CH₄ and 90.1% CH₄ with feeding rates of 60.5 and 200 L/L/day, respectively. Conversion of H_2 :CO₂ \rightarrow CH₄ was considered to be the mass based stoichiometric ratio of 8:44 \rightarrow 16. For numerical modelling of scale up it was assumed that stoichiometric conversion could be achieved at a mixed gas feed rate of 200 L / L / day with a carbon dioxide input of 14,350 m³ / day and a hydrogen input of 50,877 m³ / day.

Energy inputs into the biomethanation system comprised of:

- Mixing Energy required to maximise the solution of gases to the liquid medium where they can be utilised by the hydrogenotrophic archaea.
- 2. Heating energy Required to maintain reactor temperature at 37 °C.
- Water Removal Required to remove water generated by the stoichiometric combination of hydrogen and carbon dioxide.
- 2.4.1 Mixing Energy

Liquid within the reactor and recirculation loop were in hydraulic continuity and the static head difference was therefore assumed to be zero. The primary effort undertaken by the recirculation pump was therefore in overcoming the friction associated with the fluid flow through the system. Flow velocity was calculated and the Reynolds number determined using Equation (2) to establish whether flow was laminar or turbulent.

Equation 2: Calculation of Reynolds Number for Fluid Flow

$$Re = \frac{(U_m \times D)}{v}$$

Where:

Re = Reynolds Number

- U_m = Velocity of Flow (m / s)
- D = Pipe Diameter (m)
- v = Kinematic Viscosity (of water) (m² / s)

Construction of the reactor and the recirculation pipe was assumed to be of stainless steel with a surface roughness (k) of 0.5 μ m. This figure was used in conjunction with

the Reynolds number to read a Friction Coefficient (Cf) of 0.002 from a Moody Chart. The same value (0.0022) friction coefficient was calculated when using the Haaland formula (Equation (3)), and the Friction Head was then calculated using Darcy's Formula (Equation (4)). Friction head losses associated with 90 ° pipe bends were calculated using Equation (5). Finally, the pump power was calculated using Equation (6).

Equation 3: Calculation of friction coefficient using the Haaland equation

$$\frac{1}{\sqrt{C_f}} = -3.6 \log_{10} \left\{ \frac{6.9}{R_e} + \left(\frac{\varepsilon}{3.71} \right)^{1.11} \right\}$$

Where:

 $C_f = Friction Coefficient$

Re = Reynolds Number

 ϵ = Relative Surface Roughness (= k/D)

Equation 4: Calculation of Friction Head using Darcy's equation

$$h_f = \frac{4 \cdot C_f \cdot L \cdot v_m^2}{2 \cdot g \cdot D}$$

Where:

- $h_f =$ Friction head (m)
- $C_f =$ Friction Coefficient (calculated above)
- L = Pipe length (m)
- V_m = Mean velocity of fluid (m / s)
- $g = gravity (9.81 m / s^2)$

D = Pipe Diameter (m)

Equation 5: Calculation of friction in pipe bends

$$Hl = \left\{ k \cdot \left(\frac{v^2}{2g}\right) \right\} \cdot y$$

Where:

- Hl = Head loss due to pipe bends (m)
- K = Resistance Coefficient
- v = velocity (m / s)
- $g = Gravity (9.81 m / s^2)$
- y = Number of bends in system

Equation 6: Calculation of required Pump Power

$$P = \frac{Q \cdot H \cdot \rho \cdot g}{3.6 \times 10^6}$$

Where:

- P = Pump power (kW)
- Q = Fluid flow rate (m³ / hr)
- H = Total system differential head (m)
- $\rho = Fluid density (kg / m^3)$
- $G = gravity (9.81 m / s^2)$

A pump efficiency of 75 % was assumed to give an overall shaft power requirement to achieve the required liquid recirculation in the biomethanation reactors.

2.4.2 Heating Energy

Biomethanation reactors were assumed to operate at 37 °C. Average outside air temperature was assumed to be 10 °C. Gas input was assumed to be pre-heated (using excess heat from the electrolysers) to a temperature of 37 °C. Heat transfer coefficient for reactor walls was assumed to be $0.7 \text{ W} / \text{m}^2 / \text{°C}$ which is equivalent to heat losses from insulated tanks constructed of 300 mm thick reinforced concrete (Tchobanoglous et al., 2003). Heat losses from reactor walls, bases and tops were calculated using Equation (7).

Equation 7: Calculation of heat losses from reactors (and pipework)

$$q = U \cdot A \cdot \Delta T$$

Where:

q = Heat loss by conduction

- U = Heat transfer co-efficiency (W / m^2 / °C)
- A = Surface area of reactor vessel (m²)
- ΔT = Heat differential across surface (°C)

Heat input to bring reactors to temperature at the beginning of operation was not included as this was considered to be insignificant over the operational life of the plant.

2.4.3 Energy for Water Removal

As per Equation 1, the stoichiometric conversion of CO₂ and H₂ to CH₄ results in the creation of 2 moles of water for every mole of CH₄ produced. This water must be

removed from the reactors in order to prevent them from overfilling, ideally in such a way as to maintain the nutrient content within the reactor. For the purposes of this model, a combination of decanter centrifuge, ultrafiltration and reverse osmosis was utilised to separate biomass, nutrients and water from the removed liquid stream with solid residues being returned to the reactor and recovered liquids leaving the system. Energy inputs considered were 3.5 kWh / m³ (centrifuge), 12 kWh / m³ (ultrafiltration) and 6 kWh / m³ (reverse osmosis) (Fuchs and Drosg, 2010).

2.5 Utilisation of Oxygen

The system was modelled such that oxygen produced by the electrolysers was utilised within the conventional WWTP as an oxygenating media for secondary treatments. Flow rate into the WWTP was 180 million litres / day. BOD of the influent was assumed to be 220 mg / l and the BOD of treated water was assumed to be 20 mg / l. 1 kg of oxygen was assumed to remove 1 kg of BOD. The Oxygen Transfer Requirement to treat BOD ($O_{(req)}BOD$) was therefore 36,000 kg O₂ / day. Oxygenation of wastewater also resulted in the reduction of Total Kjeldahl Nitrogen (TKN) from an influent concentration of 40 mg / l to 20 mg / l with 4.57 kg of oxygen required to oxygenate 1 kg of TKN. The Oxygen Transfer Requirement to treat nitrogen ($O_{(req)}TKN$) was therefore 16,452 kg O₂ / day. This gave a Total Oxygen Requirement (OTR) of 52,452 kg O₂ / day (2,185 kg O₂ / hr).

The Standard Oxygen Transfer Rate (SOTR) of a diffused aeration system intended to meet the OTR was calculated using Equation (8). The air (or oxygen) volume required to meet the Standard Oxygen Transfer Rate was then calculated using Equation (9).

Finally, the power requirement for the gas blower needed to deliver either the air or oxygen to the WWTP was calculated using equation (10).

Equation 8: Standard Oxygen Transfer Rate (SOTR) of a diffused aeration system

$$SOTR = \frac{OTR}{\left\{ \left(\beta . C_{s,t} - C_w \right) / C_{s,20} \right] . \alpha F . (\theta^{T-20}) \right\}}$$

Where

- SOTR = Standard Oxygen Transfer Rate
- OTR = Field Oxygen Transfer Requirement
- β = 0.95 Cs (wastewater) / Cs (tap water)
- Cs,t = 8.19 Oxygen saturation concentration corrected for altitude and

temperature

Cw = 3.0	Operating dissolved oxygen concentration
Cs,20 = 9.02	Oxygen saturation concentration for tap water at 20 °C
αF = 0.45	Oxygen transfer efficiency ratio for fine bubble diffusers
θ = 1.024	Arrhenius constant (for temperature correction)
T = 5 °C	Outside average air temperature

Equation 9: Air (or Oxygen) requirement to meet SOTR

$$V = \frac{SOTR}{(60.\rho.SOTE.O_{cont})}$$

Where:

V = Required air (or oxygen) volume (m³)

SOTR = Standard Oxygen Transfer Rate (Equation 7)

 ρ = Density of air (or oxygen) (kg / m³)

SOTE = 0.283 Standard Oxygen Transfer Efficiency

O_{cont} = Oxygen Content of media (0.23 for air, 1.0 for oxygen)

Equation 10: Calculation of blower power requirement

$$Pw = \left(\frac{(W \cdot R \cdot T1 / hp \cdot n \cdot em)}{\{[(p_2 / p_1)^n] - 1\}} \right) \cdot ee$$

Where:

Pw = Power rating (kW)

- W = Mass flow of air (or oxygen) (kg / s)
- R = Engineering constant for air (286.9 J / kg K)
- T1 = Absolute inlet temperature (278.15 K)
- hp = Power (1000 J / s kW)
- n = SOTE (Air = 0.283) (Oxygen = 0.259)
- em = Blower mechanical efficiency (0.75)
- $p_2 = Absolute outlet pressure (kg / cm^2)$
- $p_1 = Atmospheric pressure (kg / cm^2)$
- ee = Blower electrical efficiency (0.85)

3. Results & Discussion

The numerical model described in Section 2 allowed the configuration and material flows within the **novel**, integrated system to be established (Figure 4).

The production of each additional 1.0 m³ of biomethane from the biomethanation plant (i.e. in addition to the 1.86 m³ of biomethane produced by the water scrubbing upgrading plant) would require the input of 1.19 m³ of off gas (as a CO₂ source) from the flash tank of the upgrading plant, which would be produced from the upgrading of 3.05 m³ of raw biogas from the AD plant. Hydrogen for each additional 1.0 m³ of biomethane from the biomethanation plant would require an electrolyser water input of 13.6 litres of water which would produce 0.341 kg (3.79 m³) of hydrogen with the expenditure of 18.24 kWh of electricity (approx. 77.5% of water input is unconverted). A mass of 2.72 kg (2.05 m³) of oxygen would also be available for utilisation within the WWTP. Biologically catalysing the carbon dioxide and hydrogen to methane within the biomethanation reactor would require the input of 1.99 kWh of electricity per m³ of biomethane produced – this includes reactor mixing, heating and removal of excess water.

Table 1 summarises the major material flows through the proposed full-scale system. 11 No. 1030 kW electrolysers, which would produce a total output of 50,876.5 m³ H₂ / day at constant operation would be sufficient to utilise 14,349 m³ / d of CO₂ produced by the biogas plant. Electrolysers would also produce 27,490.9 m³ of O₂ per day for utilisation within the WWTP.

The biomethanation system under consideration is assumed to be capable of converting 200 litres of feed gas (mixed H₂ and CO₂) per litre of reactor per day at ambient pressure. As such, a total working reactor volume of 326 m³ is required for the biomethanation process. In the configuration modelled this working volume was split

into 8 No. reactors, each with a working volume of 41 m³. Assuming that 98% of stoichiometric conversion can be achieved (as continuously achieved by Savvas et al, 2017a) 13,420 m³ / day of CH₄ would be produced along with approximately 20,170 litres of H₂O. If this low carbon methane was added to the gas grid it would have a market value of £1.54 million per year and would attract a further its value £1.38 million per year based on Renewable Heat Incentive (Ofgem, 2016) values at the end of 2016.

The results suggest that it would be possible in a practical sense to scale up the laboratory process described by Savvas et al (2017a) and integrate it within a large WWTP such that the plant provides a number of new and novel strategic functions, namely the biological recycling of CO₂ and the integration and balancing of power and gas grids.

3.1 Oxygen Utilisation

Approximately 36,590 kg / day (27,490 m³ / day NTP) of oxygen would be produced by the electrolysers when operating at nominal output. Based on the calculations described above, the WWTP would require an oxygen input of >1.07 million kg / day (>807,000 m³ / day) to meet its oxygenation requirements and as such the electrolysers present would only meet approximately 3.4 % of the conventional WWTPs oxygenation requirements. The reduction in power consumption by air / gas blowers would be equivalent to approximately 1,678 kWh / day, which, assuming an electricity price of £0.03 / kWh, would equate to a saving of approximately £18,383 / yr. Based on this relatively low contribution to the overall oxygenation requirement of the conventional WWTP, a decision to implement a PtM scheme would largely be promoted by the green methane generated rather than the oxygen provision. The intermittency of oxygen availability where electrolysers were to be powered by renewables would also require either additional storage or another mechanism to avoid variations in treatment conditions within the WWTP. These considerations have only been defined for a conventional WWTP reliant on active aeration requirements. Other more novel operations for sewage treatment may be possible in the future e.g. based on lower oxygen input treatments, relying on anaerobic or more passive aerobic systems such as in bioelectrochemical system (Kelly and He, 2014, Khalfbadam et al., 2016). In addition, the considerations made here were based on a narrow system boundary to utililise CO₂ from one WWTP. A more regional based approach could be possible whereby larger deployment of PtG and PtM would then yield higher oxygen levels for multiple WWTPs.

3.2 Energy Balance

Results of the energy balance of the proposed system are provided in Table 2. Results show that the electrolyser element of the system has an energetic efficiency of approximately 81.3 % providing that heat recovery is undertaken. Where no heat recovery is in place, efficiency is reduced to 62.2 %. The energy input required to operate the electrolysers at constant output is 244,728 kWh / day. The full scale biomethanation system required a total energetic input of 26,748.7 kWh / day to convert the 4,573.8 kg (50,876.5 m³ at NTP) of hydrogen and 26,432.3 kg (14,349.8 m³ at NTP) of CO₂ to 8,964.6 kg (13,420.1 m³ at NTP) of methane. This energy requirement breaks down to 25,922.6 kWh / day for mixing reactor contents, 382.2 kWh / day for reactor heating, and 443.9 kWh / day for removal of excess water. The energetic content of the methane produced is 133,825.5 kWh / day (based on 9.97 kWh / m³) giving an overall

parasitic energy demand for the biomethanation element of the process of 19.9 % of the energy produced. For this particular site conditions, expenditure of a further 855.4 kWh / day of energy would be required to compress the methane produced to 7 bar prior to grid injection. Considering the integrated power to methane system as a whole, 66.2 % of the primary energy invested would be recovered either as additional methane gas or thermal energy. In this limited system boundary comprising of a single WWTP the oxygen produced was not utilised (for the reasons described previously) and no energetic benefit was included relating to oxygen use.

As shown above and in Figure 5, by far the largest energy input is required for the electrolysis of water to produce renewable hydrogen – this accounts for 89.9 % of the total energetic input. Of the remaining 10.1 % required for biomethanation and gas compression, 9.5 % of total energetic input is required for the mixing of the liquid substrate within the reactors to maximise gas / liquid transfer. Reactor heating, removal of water using centrifugation, ultrafiltration and reverse osmosis, and final gas compression were found to have relatively minor energetic requirements (0.1 %, 0.2 % and 0.3 % of total system energy input, respectively). Figure 6 shows Sankey diagrams summarising the main mass flows (6a) and energy inputs and outputs (6b).

3.2.1 Reduction of Parasitic Load

With any energy conversion processes, there are energetic losses. In the system considered approximately 66.2 % of primary energy expended is recovered either as biomethane or thermal energy. The majority of energy (89.9 %) is expended for the electrolytic production of hydrogen. The biomethanation process as modelled had a parasitic energy demand of approximately 19.9 % of the energy content of the methane

produced, the majority of which was expended on high rate mixing with the aim of maximising the transfer of gaseous hydrogen to the liquid media. There are, however, several strategies, which could be explored in future biomethanation configurations that could significantly reduce parasitic energy demand. Operating the biomethanation process at a pressure of 10 bar, whilst requiring an energy input to reach pressure, dramatically increases the solubility of gases and helps to overcome the major rate limiting factor of the low solubility of hydrogen gas. Hydrogen is produced by the electrolysers at a pressure of 20 bar and therefore would not require further compression prior to biomethanation. To compress the CO₂ to 10 bar would require an energy input of 0.04 kWh / kg CO₂ input (0.08 kWh / m³ CO₂ NTP), however, the increased gasliquid transfer means that a far smaller reactor would be required (32.6 m³ instead of 326 m³) which in turn would reduce the mixing energy required from 25,922 kWh / day to 1,467.3 kWh / day. This could reduce the parasitic energy demand of the biomethanation process from 19.9% to <5% of the energy content of the methane produced, assuming that there is no physiological impediment to the microbial population converting the gas supplied.

The high mixing requirement of gas/liquid reactors such as that modelled in this study also provides practical problems. The ability of pumps to entrain significant volumes of gas within a liquid stream is limited, and, as such, alternative reactor designs that utilise minimal volumes of liquid such as that described by Savvas et al. (2017b) would be both practically and energetically beneficial providing that methane yields equivalent to, or greater than, those modelled in this study can be achieved.

3.3 Process Economics and Regulatory / Policy Support

From an economic perspective, the benefits of the process are not easily quantifiable without a supporting legislative framework that recognises the value in flexibly managing interlinked gas and electricity networks. Specifically, excess renewable electricity that would currently be curtailed could be made available at lower than standard cost. The process requires an input of 272.3 MWh / day of electricity and in order to deliver reasonable payback times this input electricity would have to cost approximately 1.3 p / kWh, which might be feasible where grid balancing / constraint reduction services are being delivered. A future pricing structure would also need to reflect the functions delivered by the technology of conversion of energy vectors, the ability to store energy and the associated grid management benefits, and this is likely to require financial income approximately equivalent to the current UK Renewable Heat Incentive (RHI) value (4.32 - 1.96 p / kWh according to the amount exported). The value of the methane produced is approximately $\pounds 1.54$ million per year. Supplying this renewable gas to the grid would currently attract an additional £1.38 million through RHI payments. The approximate economic performance of the technology based on these assumptions is shown in Table 3 As concluded by Walker et al. (2016) it is clear that a future pricing structure of any PtG (including P2M) must assign value to the various functions that such a system would deliver, including:

- Linking electricity networks to gas grids to allow grid scale storage of renewable energy.
- Maximising output (and therefore economic income) of existing renewable electricity assets by reducing curtailment events at times of low electricity demand and high renewable energy production.

- Enabling the development of renewable energy assets in areas with high renewable energy availability but severe electricity grid constraint, or no electricity grid at all (i.e. maximising the deployment and generation of renewable electricity)
- 4. Increasing the supply of 'low carbon' gas to the gas networks, and prolonging the operational viability of gas grids.
- 5. Utilising and valorising industrial carbon dioxide emissions.

These are all strategic issues of national and international significance that, if successfully addressed, would bring widespread economic and environmental benefits to society. A regulatory and financial framework that recognises the potential benefits of this, or other technologies that could bring similar benefits, is therefore urgently required at regional, national and international levels. Whilst this study reflects the applicability of the technology for a one WWTP only and in isolation with other systems, PtG and PtM applications on a more regional and national levels could provide additional economic and environmental benefits.

4. Conclusions

Biomethanation of CO₂ from biogas produced at a WWTP, with hydrogen produced from electrolysis of water, would provide a regionally significant link between electricity and gas grids. The hydrogen production and biomethanation system had an overall efficiency of 66.2 %. The un-optimised biomethanation system had a parasitic energy requirement of 19.9 % of energy produced, with strategies proposed to reduce this to <5 %. To achieve financial viability the strategic industrial, environmental and economic benefits of technologies that integrate electricity and gas grids must be recognised within policy, regulatory and financial frameworks that reflect the services and benefits provided.

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References

Bassani, I., G. P. Kougias, L. Treu and I. Angelidaki (2015). "Biogas Upgrading via Hydrogenotrophic Methanogenesis in Two-Stage Continuous Stirred Tank Reactors at Mesophilic and Thermophilic Conditions." <u>Environmental Science and Technology</u> **49**(20): 12585-12593.

BEIS (2016). Digest of United Kingdom Energy Statistics 2016. Available at https://www.gov.uk/government/uploads/system/uploads/attachment_data/file/577712/ DUKES_2016_FINAL.pdf. Accessed on 16th February 2017, Department for Business, Energy and Industrial Strategy.

Bensmann, A., R. Hanke-Rauschenbach, R. Heyer, F. Kohrs, D. Benndorf, U. Reichl and K. Sundmacher (2014). "Biological methanation of hydrogen within biogas plants: A model-based feasibility study." <u>Applied Energy</u> **134**: 413-425.

Carmo, M., L. D. Fritz, J. Mergel and D. Stolton (2013). "A comprehensive review on PEM water electrolysis." <u>International Journal of Hydrogen Energy</u> **38**: 4901-4934.

Fuchs, W. and B. Drosg (2010). Technologiebewertung von Gärrestbehandlungs – und Verwertungskonzepten. Universität für Bodenkultur Wien ISBN 978-3-900962-86-9.

Götz, M., J. Lefebvre, F. Mörs, M. A. Koch, F. Graf, S. Bajohr, R. Reimert and T. Kolb (2016). "Renewable Power-to-Gas: A technological and economic review." <u>Renewable Energy</u> **85**: 1371-1390.

Gutiérrez-Martín, F. and M. L. Rodríguez-Antón (2016). "Power-to-SNG technology for energy storage at large scales." <u>International Journal of Hydrogen Energy</u> **41**: 19290-19303.

Hirth, L., F. Ueckerdt and O. Edenhofer (2015). "Integration costs revisited - An economic framework for wind and solar variability." <u>Renewable Energy</u> **74**: 925-939.

HSE (2015). Injecting hydrogen into the gas network - a literature search. Available at http://www.hse.gov.uk/research/rrpdf/rr1047.pdf. Accessed 6th February 2017, The Health and Safety Executive.

Jensen, H. S., H. P. Larsen and M. Mogensen (2007). "Hydrogen and synthetic fuel production from renewable energy sources." <u>International Journal of Hydrogen Energy</u> **32**: 3253-3257.

Jürgensen, L., A. Ehimen, E, J. Born and B. J. Holm-Neilson (2015). "Dynamic biogas upgrading based on the Sabatier process: Thermodynamic and dynamic process simulation." <u>Bioresources Technology</u> **178**.

Kelly, P. T. and Z. He (2014). "Nutrients removal and recovery in bioelectrochemical systems: A review." <u>Bioresource Technology</u> **153**: 351-360.

Khalfbadam, H. M., M. P. Ginige, R. Sarukkalige, A. S. Kayaalp and K. Y. Cheng (2016). "Bioelectrochemical system as an oxidising filter for soluble and particulate organic matter removal from municipal wastewater." <u>Chemical Engineering Journal</u> **296**: 225-233.

Leeds City Gate (2016). h21 Leeds City Gate. http://www.northerngasnetworks.co.uk/wp-content/uploads/2016/07/H21-Report-Interactive-PDF-July-2016.pdf.

Luo, G. and I. Angelidaki (2012). "Integrated Biogas Upgrading and Hydrogen Utilization in an Anaerobic Reactor Containing Enriched Hydrogenotrophic Methanogenic Culture." <u>Biotechnology and Bioengineering</u> **109**: 2729-2736.

Luo, G., S. Johansson, K. Boe, L. Xie, Q. Zhou and I. Angelidaki (2012). "Simultaneous Hydrogen Utilization and In Situ Biogas Upgrading in an Anaerobic Reactor." <u>Biotechnology and Bioengineering</u> **109**: 1088-1094.

Malmberg Water AB (2015). Off gas specification for Malmberg water scrubbing system. Personal correspondence received by S. Esteves on 16/06/15.

Martin, R. M., J. F. Jeffrey, R. Stark, L. Mets and L. Angenent (2013). "A Single-Culture Bioprocess of *Methanothermobacter thermautotrophicus* to Upgrade Digester Biogas by CO2-to-CH4 Conversion with H2." <u>Archaea</u> 2013.

Ofgem (2016). Non-Domestic Renewable Heat Incentive (RHI). Available at https://www.ofgem.gov.uk/environmental-programmes/non-domestic-rhi, Ofgem,.

Reiter, G. and J. Lindorfer (2015). "Global warming potential of hydrogen and methane production from renewable electricity via power-to-gas technology." <u>International</u> Journal of Life Cycle Assessment **20**(4): 477-489.

Savvas, S., J. Donnelly, T. Patterson, Z. Chong, S and P. S. Esteves (2017b). "Biological methanation of CO2 in a novel biofilm plug flow reactor: A high rate and low parasitic energy process." <u>Applied Energy</u> **202**: 238-247.

Savvas, S., J. Donnelly, T. Patterson, R. Dinsdale and S. Esteves (2017a). "Closed Nutrient Recycling via Microbial Catabolism in an Eco-Engineered Self Regenerating Mixed Anaerobic Microbiome for Hydrogenotrophic Methanogenesis." <u>Bioresources Technology</u> **227**: 93-101.

Sternberg, A. and A. Bardow (2016). "Life Cycle Assessment of Power-to-Gas: Syngas vs Methane." <u>Sustainable Chemistry and Engineering</u> **4**(8): 4156-4165.

Tchobanoglous, G., L. F. Burton and H. D. Stensel (2003). <u>Wastewater Engineering:</u> <u>Treatment and Reuse / Metcalf & Eddy Inc.</u>, McGraw-Hill.

UK Government (1996). Gas Safety (Management) Regulations. Available at http://www.legislation.gov.uk/uksi/1996/551/regulation/1/made, Crown Copyright.

Van den Bergh, K., D. Couckuyt, E. Delarue and W. D'haeseleer (2015). "Redispatching in an interconnected electricity system with high renewables penetration." <u>Electric Power Systems Research</u> **127**: 64-72.

Vo, T. Q. T., A. Xia, M. D. Wall and D. Murphy, J (2017). "Use of surplus wind electricity in Ireland to produce compressed renewable gaseous transport fuel through biological power to gas systems." <u>Renewable Energy</u> **105**.

Walker, B. S., D. van Lanen, M. Fowler and U. Mukherjee (2016). "Economic analysis with respect to Power-to-Gas energy storage with consideration of various market mechanisms." <u>International Journal of Hydrogen Energy</u> **41**: 7754-7765.

Zoss, T., E. Dace and D. Blumberga (2016). "Modeling a power-to-renewable methane system for an assessment of power grid balancing options in the Baltic States' region." <u>Applied Energy</u> **170**: 278-285.

Figures and Tables

Figures



Figure 1 – Scope and Boundary of the System Considered in the Feasibility Study



Figure 2 – Schematic of major elements of the conventional WWTP considered in the

<u>study</u>



Figure 3 – Schematic Showing the Primary Elements of the Biomethanation Process







Figure 5 – Breakdown of energetic input to the Power to SNG system

Electrolyser Water Input: 182 952	Unconverted Water: 141 7	88		6a
		Oxygen: 36 590		Excess Carbon Dioxide: 1 276 Methane Produced: 8 965
Carbon Dioxide: 26 431	Hydrogen: 4 5/4	Biometh. Mass Input: 31 005		Water Production: 20 170
				Unconverted Carbon Dioxide: 503 Unconverted Hydrogen: 91
Primary Energy: 272 332		Electrolysis stage energy loss: Non Recoverable Heat:	26 071 19 905	6b
	Electrolyser Heat Production: 66 350			Recoverable Heat: 46 444
	Electrolyser Input Energy: 244 728 Chemical Energy of Hydrogen: 152 307	Biomethanation Input Energy	: 179 056	Chemical Energy of Methane: 133 826
	Biometh. Mixing: 25 923 — Biometh. Dewatering: 444			

Figure 6 – Sankey Diagram of (6a) Mass Balance (kg / d) and (6b) Process Energy

Inputs and Outputs (kWh / d)

Tables

Table 1 – Major Material Flows of Scaled Up Power to Methane System

Plant Component / Material	Plant Description / Material Volume
Biogas Plant	
Volume of CO ₂ Produced	14,720 m ³ / day
Electrolyser	11 No. 1030 kW
Volume of H ₂ O Utilised	182,9521/day
Volume of H ₂ Produced	50,876.5 m ³ / day (NTP)
Volume of O ₂ Produced	27,490 m ³ / day (NTP)
Biomethanation Plant	Mesophilic reactors, ambient pressure, receiving 2001/1/
	day of mixed H ₂ and CO ₂
Volume of CO ₂ utilised	14,349 m ³ / day (NTP)
Mixed Gas Input Volume	65,226.3 m ³ / day (NTP)
Total Working Reactor Volume	326 m^3
Required	
Individual Reactor Working	41 m ³
Volume	
Number of Reactors Installed	8 No.
Volume of CH ₄ Produced	$13,420.1 \text{ m}^3 / \text{day} (\text{NTP})$
Volume of H ₂ O Produced	20,170.5 1 / day

1 able 2 – Modelled Energy Balance of the Integrated Syste

Electrolyser		
Primary Energy Input (Renewable / Grid Elec.)	244,728 kWh / day	
Hydrogen Output (Mass)	4,573.8 kg / day	
Hydrogen Energy Content (LHV)	152,307 kWh / day	
Oxygen Output (Mass)	<mark>36,590.4</mark> kg / day	
Waste Heat Production	66,349 kWh th / day	
Estimate of recoverable heat energy (70%)	<mark>46,444</mark> kWh th / day	
Overall Electrolyser Efficiency (with heat recovery)	81.2 %	
Overall Electrolyser Efficiency (no heat recovery)	<mark>62.2 %</mark>	
Biomethanation		
Reactor Heating Energy	382.2 kWh / day	
Mixing Energy	25,922.6 kWh / day	
Water Removal Energy	443.9 kWh / day	
Total Energy Input	26,748.7 kWh / day	
Volume of methane produced	13,420.1 m ³ (NTP)	
Energy expended per m ³ CH ₄ output	1.99 kWh / m ³ CH ₄	
Produced Methane Energy Content	133,825.5 kWh / day	
Biomethanation parasitic energy demand	19.9 % of produced methane	
Compression Energy (for grid injection)		
Compression of Produced CH ₄ (7 bar)	855.4 kWh / day	
Overall Power to Methane System Efficiency		
Electrolyser primary energy input	244,728 kWh / day	
Biomethanation energy input	26,748.7 kWh / day	
Compression energy input	855.4 kWh / day	
Total Energy expenditure	272,332.1 kWh / day	
Heat recovery from electrolyser	46,444 kWh th / day	
Energy content of methane produced	133,825.5 kWh	
Total energy produced / saved	180,270.4 kWh / day	
% of primary energy recovered / saved	66.2 %	

Table 3 – Estimated Economic Parameters for the Proposed Process

Description	Current Cost Estimate	Future (circa + 5 yrs) Cost
		Estimate
<mark>Capital Costs</mark>		
Electrolysers	£7,500,000	£6,200,000
Biomethanation Plant	£1,300,000	£1,100,000
Installation Costs	£600,000	£500,000
Total	£9,400,000	£7,800,000
Operation and Maintenance		
Energy Input	<mark>99,401 MWh / yr</mark>	<mark>91,340 MWh / yr</mark>
<mark>Cost (per kWh)</mark>	£13 / MWh	£13 / MWh
Energy Cost	£1,292,213 / yr	£1,187,420 / yr
Operator Allowance	£35,000 / yr	£35,000 / yr
Maintenance Allowance	£437,580 / yr	£361,000 / yr
Total O&M Cost	£1,764,793 / yr	£1,583,420 / yr
Additional Income		
Energy Content of Additional		
Methane	<mark>43,961 MWh / yr</mark>	<mark>43,961 MWh / yr</mark>
Energy Value of Methane	£35 / MWh	£35 / MWh
Value of Additional Methane	£1,538,635 / yr	£1,538,635 / yr
RHI (or future equivalent)	£1,386,628 / yr	£1,386,628 / yr
Total Income	£2,925,263 / yr	£2,925,263
Interest Rate	<mark>4 %</mark>	<mark>4 %</mark>
Payback Time	10 yrs	<mark>6.5 yrs</mark>