Techno-economic analysis of biogas upgrading via amine scrubber, carbon capture and ex-situ methanation

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Abstract

Biogas upgraded to biomethane can provide a renewable gaseous transport fuel and is one of the proposed solutions in meeting the renewable energy supply in transport targets set under the EU Renewable Energy Directive. The upgrading process for biogas involves the removal of CO_2. Amine scrubbing is one traditional method of upgrading that is applied due to its low methane slippage and its capability to provide a high purity renewable methane product. However, new technologies such as power to gas (P2G) can also upgrade biogas through biological methanation by combining the CO_2 in biogas with H_2 to produce renewable methane. The H_2 for P2G can be produced through electrolysis of renewable electricity. Through simulation software – SuperPro Designer, the economics of different pathways for upgrading biogas from a grass silage and slurry fed digester are analysed and compared in this paper. Three scenarios are investigated: biogas upgrading through amine scrubbing (scenario 1); biogas upgrading through amine scrubbing with CO_2 directed to ex-situ biological methanation (scenario 2) and biogas upgrading through ex-situ biological methanation only (scenario 3). The results show that at a net present value of zero, the minimum selling price (MSP) per m^3 of renewable methane for scenario 1, 2 and 3 is €0.76; €1.50 and €1.43, respectively (with an electricity price to produce H_2 of €0.10/kWh and a grass silage production cost of €27/t). The electricity price has a significant effect on the cost of renewable methane in both scenarios 2 and 3. The MSP reduces to €1.09 and €1.00 per m^3 of renewable methane, respectively for scenarios 2 and 3, if the electricity price is reduced to €0.05/kWh. Since the renewable methane MSP from scenario 2 is higher than scenario 3, it is suggested that direct biogas injection to the methanation reactor is financially more attractive than capturing CO_2 from biogas and feeding it to the methanation step. The MSP of renewable methane from both scenarios 2 and 3 are significantly higher than that of scenario 1. However, when considering climate change mitigation, balancing of the electricity network and storage of surplus electricity, utilising P2G can offset some of these costs. If considering methanation as the upgrading unit of a biogas plant, its fixed capital cost is approximately the same as that of the amine scrubber. The cost of H_2 is a significant factor in determining the cost of renewable methane.

Keywords: CO_2 capture; process simulation; amine scrubber; methanation; biological Power to Gas; economic analysis

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Abbreviations

CSTR: Continuously stirred tank reactor

DS: Dry solids

HRT: hydraulic retention time

MEA: mono-ethanol-amine

MSP: minimum selling price

NPV: Net present value

P2G: Power to gas

S1: Scenario 1

S2: Scenario 2

S3: Scenario 3

t: tonne

VS: Volatile solids

wwt: wet weight ton

Mm^3: Million m^3

Mt: Million tonnes

1. Introduction

A simplified breakdown of energy consumption in Ireland is ca. 20% electricity, 40% heat and 40% transport [1]. The EU 2020 targets for renewable energy is 16% with a national breakdown in each of those sectors set at 40%, 12% and 10%, respectively [2]. The production of renewable electricity has primarily been the focus in Ireland and as a result, its production has accelerated beyond renewable heat and transport. Anaerobic digestion of available biomass to generate renewable methane is one pathway that has been identified for potentially contributing to both the transport and heat sectors. For instance, grass is the most important agricultural crop in Ireland; over 90% of agricultural land is covered by grass [3]. Excess grass, surplus to livestock requirements, may provide a potential feedstock for renewable methane production in Ireland [4]. Grass is deemed a promising feedstock due to its high yield, low production energy input and potentially high methane yields [5]. Previous studies have indicated that in order to have sufficient nutrients for long term digestion, grass silage should be co-digested with slurry [6]. Dairy slurry is also an abundant resource in Ireland with an annual production of ca. 7 Mt [6].

Ireland, an island state with limited electrical interconnectivity, is also expected to spill between 7-14% of its renewable electricity production by 2020, as supply will periodically exceed demand [7]. Thus, a method of storing surplus electricity is crucial. One potential method of storage is to change the energy vector from electricity to gas. This can be done through electrolysis, utilising the surplus electricity to split water into hydrogen (H_2) and oxygen (O_2) (Eq. 1). Currently, three electrolysis technologies are used for producing H_2. The alkaline electrolyser is at commercial stage, the polymer electrolyte membrane is at pilot stage and the solid oxide electrolysis cell (SOEC) is in a research and development stage [8]. A further methanation step, combining the H_2 with carbon dioxide (CO_2), is required to produce methane (CH_4) in a Sabatier reaction. These combined processes are known as Power to Gas (P2G). The equation for production of CH_4 from CO_2 and H_2 is expressed in Eq. 2.

$2H_2O(l) \rightarrow 2H_2(g) + O_2(g)$ $\Delta Hr = 286$ kJ/mole (at 25° C, 1 bar) Eq. 1

$CO_2 + 4H_2 \leftrightarrow CH_4 + 2H_2O$ $\Delta H = -165$ kJ/mole Eq. 2

The methanation step can be carried out through chemical or biological means [9]. Chemical methods use a catalyst for methanation. This is a mature technology and nickel has often been chosen as the catalyst due to its high activity and low price. The efficiency of catalytic methanation is between 70% to 85% [10]. In contrast, biological methanation uses hydrogenotrophic methanogenic archaea to consume H_2 and CO_2 as energy sources and produce CH_4 [9]. The CO_2 source can be extracted from industry through carbon capture; or from biogas plants as biogas typically contains ca. 45% CO_2. The latter is of particular interest as methanation of CO_2 and H_2 may potentially provide a form of biogas upgrading for biogas plants, offsetting costly traditional upgrading methods such as amine scrubbers. The operation of biological methanation can be in-situ or ex-situ. The in-situ method feeds H_2 directly into an anaerobic digester, while the ex-situ method uses an external reactor for the methanation step, reacting the biogas (including for CO_2) and H_2. In terms of output, in-situ biological methanation can achieve a final CH_4 content of up to 75%, whereas ex-situ can reach up to 98%, a quality similar to that of natural gas [9]. The CH_4 produced from P2G systems is equivalent to biomethane and can again be used as fuel in the heat and transport sectors. P2G technologies have generated much interest of late and have previously been reported as a more sustainable energy storage method as compared to other large scale storage technologies such as pumped hydroelectric storage and compressed air energy storage [8]. However, P2G systems will not always have a supply of surplus electricity at a cheap price to produce H_2, thus, sometimes it may be necessary to combine traditional biogas upgrading with methanation. In this case, when electricity prices are cheap (when supply exceeds demand), the electricity can be used to produce H_2 for methanation and when electricity demand is high the biogas can be upgraded by traditional upgrading.

Biogas from a biogas plant can be upgraded by water scrubbing, organic solvent scrubbing, amine scrubbing, pressure swing adsorption or by a membrane [11]. To the authors' knowledge, there is little research on techno-economic analyses of upgrading of biogas by methanation processes. Previous studies have compared the costs of CH_4 production from traditional biogas upgrading with CO_2 capture for methanation [12-14], however these studies only considered catalytic methanation. Vo *et al*. [9] calculated the costs of renewable methane from different types of feedstocks with upgrading of biogas provided by biological methanation. However, the study did not consider the capture and utilisation of CO_2 from the biogas plant. Amine scrubber technology was chosen as the upgrading methodology in this study because of its low methane loss (<0.1%) [11], in addition to this, the CO_2 from the amine scrubber could be captured and reused.

This is the first paper to undertake a techno-economic analysis of biogas upgrading comparing amine scrubbing with or without carbon capture and ex-situ biological methanation. No previous works have compared carbon capture from amine scrubbing integrated with biological methanation. Superpro Designer (Intelligen Inc., Scotch Plain, NJ, V10) was used to develop the process models.

The aim of this study is to investigate the economic viability (in terms of € per m^3 renewable methane produced) of three pathways for upgrading biogas to renewable methane generated from a typical biogas plant digesting grass silage and slurry. The three scenarios are as follows:

Scenario 1: Biogas production + amine scrubbing: The plant produces biogas (CH_4 and CO_2) from grass silage and dairy slurry, which was then upgraded by amine scrubbing to remove CO_2. This scenario did not consider CO_2 capture; CO_2 is emitted upon methane upgrading.

Scenario 2: Biogas production + amine scrubbing + ex-situ biological methanation: The plant produces biogas (CH_4 and CO_2) from grass silage and dairy slurry, which was then upgraded by amine scrubbing to remove CO_2. The CO_2 removed was captured and sent to ex-situ biological methanation in which H_2 and CO_2 was reacted to form CH_4.

Scenario 3: Biogas production + ex-situ biological methanation: Unlike other scenarios, the biogas from grass silage and dairy slurry was directly transferred to the ex-situ biological methanation system, where CO_2 in the biogas was reacted with H_2 to form CH_4.

The objectives of this study are as follows:

- Simulate and develop the three biogas upgrading scenario processes: amine scrubber; amine scrubber and ex-situ biological methanation; and ex-situ biological methanation;
- Calculate costs of renewable methane from grass silage and dairy slurry biogas plants with the three different upgrading technology scenarios;
- Analyse and compare the energy consumption of the biogas plant, amine scrubber and methanation elements;
- Estimate the effects of electricity cost and grass silage cost on the cost of renewable methane.

2. Methodology

2.1. Process development and description

The simulation was separated into two parts: i) biogas production, which includes feedstock processing, digester operation and digestate handling and ii) gas upgrading or utilization using amine scrubber and/or methanation reactor. The electrolysis step (producing H_2) was excluded from the analysis, however the cost of electricity, which affects the cost of H_2, was analysed to assess the effect of scale in the costs associated with converting H_2 to CH_4. The renewable methane produced is assumed to be compressed and injected to the distribution gas grid at 7 bar. As indicated, all three scenarios included for the same biogas plant but had a different upgrading method (Figure 1). The simulation files are attached as supplementary information to facilitate the reproduction of the results, while the screenshot of different scenarios is available from the Appendix A. The assumptions of the process are reported in Table 1.

The biogas section was simulated as in Figure 2. For the purposes of the study, the grass silage feedstock was stored in a silo pit (P-8/SL- 101), with sufficient storage capacity for a year's supply. Dairy slurry was considered to be stored in sealed, water – tight and reinforced concrete container (P-3/V- 101) for two days. The silage is shredded (P-2/SR - 101) and mixed with dairy slurry before being loaded to the digester, while the pump (P-9/PM-101) functions as both a pump and a mixer. The characteristics of the grass silage and dairy slurry (Table 2) are adopted from Wall *et al.* [6]. The ratio of grass silage and dairy slurry fed to the biogas section was calculated on a volatile solids (VS) basis; 80%VS from grass silage and 20%VS from dairy slurry. Grass silage benefits from co-digestion with dairy slurry in maintaining an optimal C:N ratio and as a source of micro-nutrients [6]. Both grass silage and dairy slurry contain nutrients, nitrogen, phosphorous and potassium [15] which are important factors in order to utilise the digestate as a fertiliser.

Table 1: Assumptions made for the three scenarios

Items	Assumptions	Note
Process	Continuous (24 hours/day for 330 days per year (7,920h))	
Grass silage (wwt/h)	3.2	
Dairy slurry (wwt/h)	3.5	
HRT at CSTR1 (days)	40	
HRT at CSTR2 (days)	30	
Project life time	20 years	
Construction period	1 year	
Calculation year	2016	
Start-up period	6 months	
NPV for three scenarios	0	At MSP, NPV is 0
Interest rate	8%	
Income tax	12.5%	
Start-up capital	No start-up capital is required from the Government	[16]
Mono-ethanol-amine	€1,500/t	
Grass silage feedstock	€27/wwt	[17]
Electricity cost for running process	€0.15/kWeh	[18]
Electricity cost for producing H_2	€0.10/kWeh	

2.1.1. Biogas section

The grass silage/slurry mixed feedstock, now with a dry solids (DS) content of 18%, is pumped to two CSTRs (in series). The two CSTRs were assumed to be operating at a mesophilic temperature range of 38°C. The HRT was set at 40 days in the first CSTR (P-6/AD-

101) and 30 days in the second CSTR (P-7/AD-102). It is assumed that 80% destruction of VS takes place in the first CSTR, with 20% of remaining VS removed in the second CSTR. In total, 84% of VS is destroyed in the process. The concentrations of CH_4 and CO_2 generated are based on the Buswell equation shown in Eq.3 [19]:

$$C_nH_aO_b + \left(n - \frac{a}{4} - \frac{b}{2}\right)H_2O \rightarrow \left(\frac{n}{2} + \frac{a}{8} - \frac{b}{4}\right)CH_4 + \left(\frac{n}{2} - \frac{a}{8} + \frac{b}{4}\right)CO_2 \qquad \text{Eq.3}$$

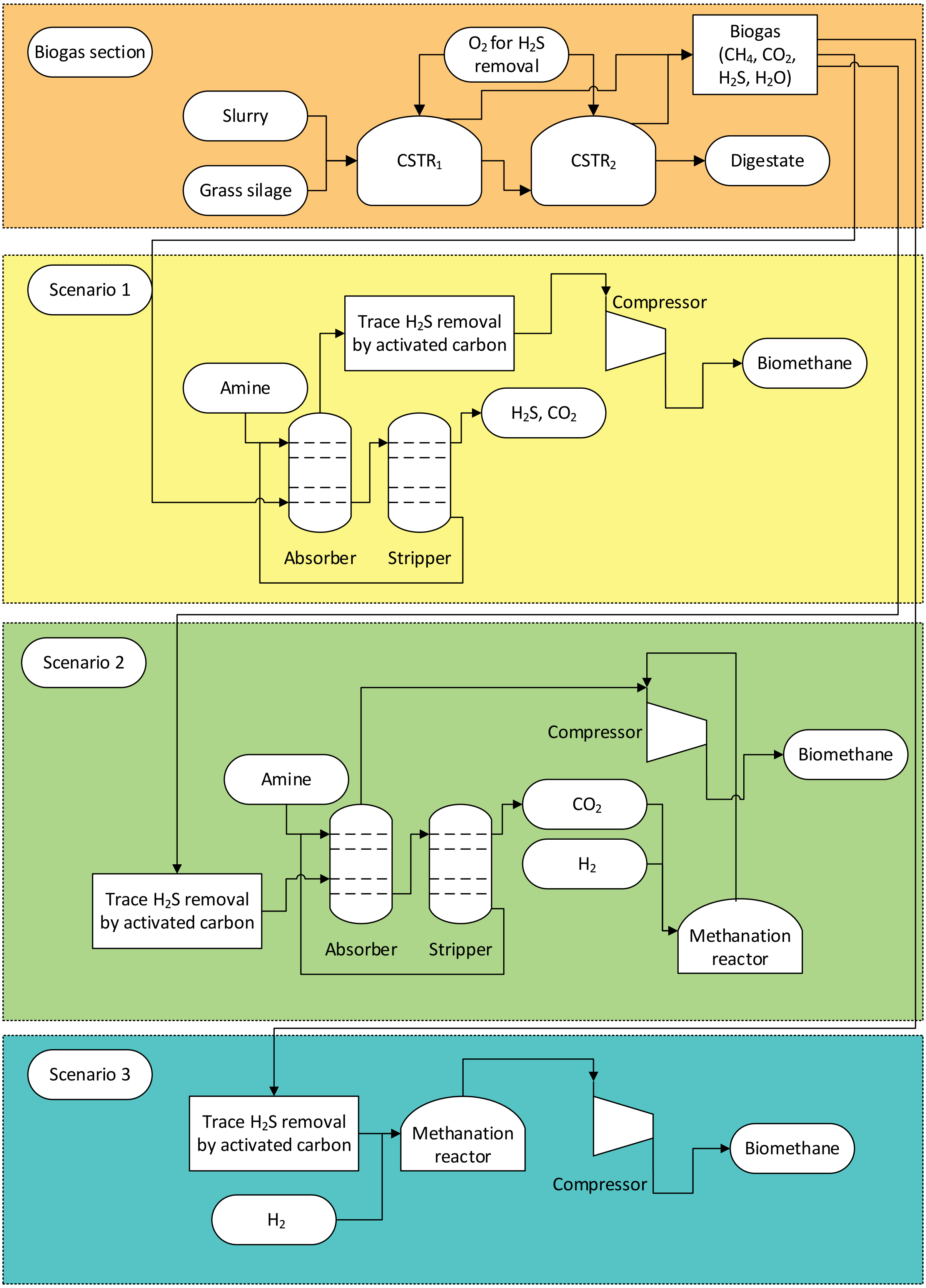


Figure 1: Process flow diagram of three biogas-upgrading methods. Scenario 1 corresponds to upgrading the biogas via amine scrubbing, while Scenario 2 corresponds to biogas upgrading via amine scrubbing and methanation reactor and Scenario 3 refers to biogas upgrading via methanation reactor.

Grass silage does not contain sulphur, while dairy slurry has 0.4 kg total sulphur per t fresh weight (Table 2). Under anaerobic conditions, sulphate is converted into hydrogen sulphide (H_2S) by sulphate – reducing bacteria. Sulphur in feedstocks containing protein when digested may also form H_2S [20]. H_2S causes corrosion to equipment and pipes when injected to the grid; it can also be toxic and inhibit anaerobic processes [21]. For injection to the gas grid or to use as gaseous fuel, the concentration of H_2S must be below 5mg/m^3 [22, 23]; an important step of biogas upgrading is eliminating H_2S. There are many methods to remove H_2S from biogas such as biological desulphurisation, iron chloride, impregnated activated carbon, iron hydroxide or oxide, and sodium hydroxide scrubbing [24]. H_2S can also be removed by combining the removal of CO_2 by pressurised water scrubbing and amine scrubbing [25]. Thus, for scenario 1, H_2S was removed with CO_2 by the amine scrubber (see section 2.1.2) as the CO_2 in this scenario is not otherwise utilised. Scenarios 2 and 3 need a separate component in order to eliminate H_2S in the methanation step. Among those methods, biological desulphurisation was chosen due to its simplicity, efficiency and low cost. In such a method, small amounts of air or O_2 (0.3 – 3%) (S-106) are added into the headspace of the CSTRs; under micro-aerobic environments, sulphide-oxidizing bacteria oxidize sulphide to elemental sulphur [21]. If air is added to the headspace, nitrogen will also be added to the biogas as a consequence, thus pure O_2, a by-product from the electrolysis process was assumed to be utilised for biological desulphurisation. Research has shown little potential for explosion; two of ten studies found a slightly lower specific methanogenic activity in micro-aerobic reactors compared to anaerobic reactors [21]. The removal efficiency of H_2S for biological desulfurization process and amine scrubbing is 88% [21] and 98% [26] respectively, thus another step should be added to reduce H_2S to 5ppm. A desulphurization process via activated carbon (Siloxa company) was chosen to reduce H_2S levels below 1ppm with a technology cost of €0.01/m^3 biogas [27].

Table 2. Characteristics of substrates (%) and nutrient concentrations kg/t fresh weight (adopted from [6] and [15]).

Substrate	Chemical formula	Dry solid (DS) (%)	Volatile solid (VS) (%)	Fixed solids or ash (%)	Water (%)	Total N	P	K	S
Grass silage	$C_{30}H_{50}O_{23}$	30	28	2	70	4	0.5	-	-
Dairy Slurry	$C_{22}H_{34}O_{19}$	8.8	6.7	2.1	91.2	3	0.5	2.9	0.4

The CH_4 product was assumed to contain some water vapour; two methods are appropriate to remove water from the gas: (i) gas dehumidification and (ii) ground tube dewatering [28]. To reduce the energy consumption, ground tube dewatering was applied in this work.

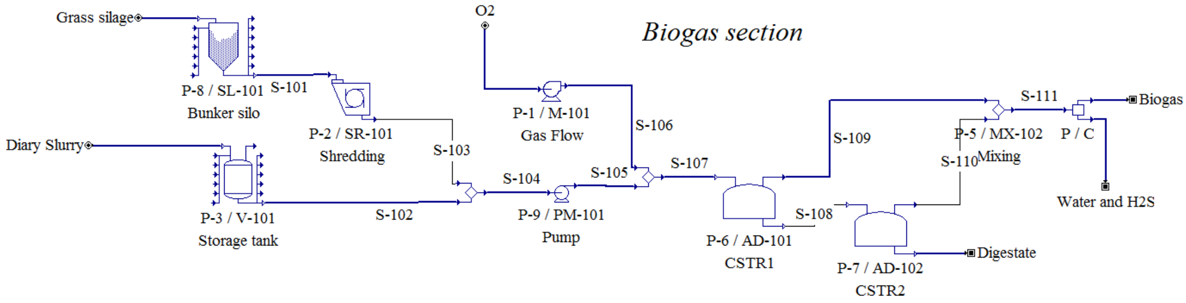


Figure 2: Biogas section simulation

2.1.2 Upgrading by amine scrubber and biological methanation

An amine scrubbing process is composed of the following elements: an absorber column; a heat exchanger; a stripper; a condenser and a re-boiler. The role of the re-boiler is to heat up the incoming amine liquid solvent and vaporise the CO_2 to obtain a lean stream of solvent. In this case (Figure 3), a blower (P-20/M-102) at the bottom introduces biogas into the absorber (P-4/C-101) and amine solution (S-112) is fed at the top of the absorber. The biogas flows up the column and the solution flows counter-currently down the column [26]. Plates or packing can provide additional surface area increasing the contact time between the gas and the solution. At the bottom of the absorption column, the solution is saturated by CO_2 and H_2S, the saturated solution is called rich amine (S-113) (the stoichiometric loading ratio is of 0.5 mole CO_2 per mole amine) [29, 30]. CH_4 (S-120) is discharged at the top of absorber and compressed to inject into the gas grid. The stripper (P-10/C-102) acts as a regeneration column whereby the rich amine solution (S-114) is heated by steam (S-118) from the re-boiler (P-15/HX-103). At a temperature of ca. 120°C, CO_2 is liberated into a concentrated stream and exits at the top of the stripper (S-115) [31]. However, not all of the CO_2 becomes free of the amine solution (called lean amine). The lowest energy requirement of 176 kJ/mole CO_2 (4 GJ/t CO_2) can be achieved at lean solvent loading between 0.25 and 0.30 mole CO_2/ mole mono-ethanol-amine (MEA) [29, 30, 32]. The CO_2 in the stripper is vaporised with water, thus this gaseous phase needs to be condensed (P-19/CSP-104) to separate water that is recycled back (S-117) to the stripper. The high temperature lean amine solution (the solution from the stripper) goes through a heat exchanger (P -17/HX-104) with the lower temperature rich amine solution. Lean amine is cooled down (P-18/HX-105) once more before going back to the absorption column, while rich amine is fed into the stripper [26]. The requirements of upgraded renewable methane (CH_4) for gas grid injection and as a potential vehicle fuel is typically >96% volume CH_4 (less than 4% volume CO_2) [33]. Amine scrubbers can provide low methane slippage (0.1%) and a high purity renewable methane end product [11]. CO_2 is emitted to atmosphere if CO_2 is not utilised in a further step; if CO_2 is used for methanation, it is injected into methanation reactor.

In the biological methanation upgrading section, the CO_2 source can come from the CO_2 stream produced as a result of biogas upgrading with amine (scenario 2), or by utilising the biogas itself directly (CH_4 and CO_2) produced from the biogas section (scenario 3). H_2 and CO_2 are assumed to be consumed biologically by hydrogenotrophic methanogenic archaea to produce CH_4 and water (Figure 3). The biological methanation reactor operates at 60˚C, ambient pressure [34] and the ideal ratio of CO_2 and H_2 is 1:4 [35]. The parasitic energy demand for stirring was suggested between 0.1 -1 kWh per m^3 methane produced [36], this study assumes this parasitic energy is 1 kWh per m^3 renewable methane produced.

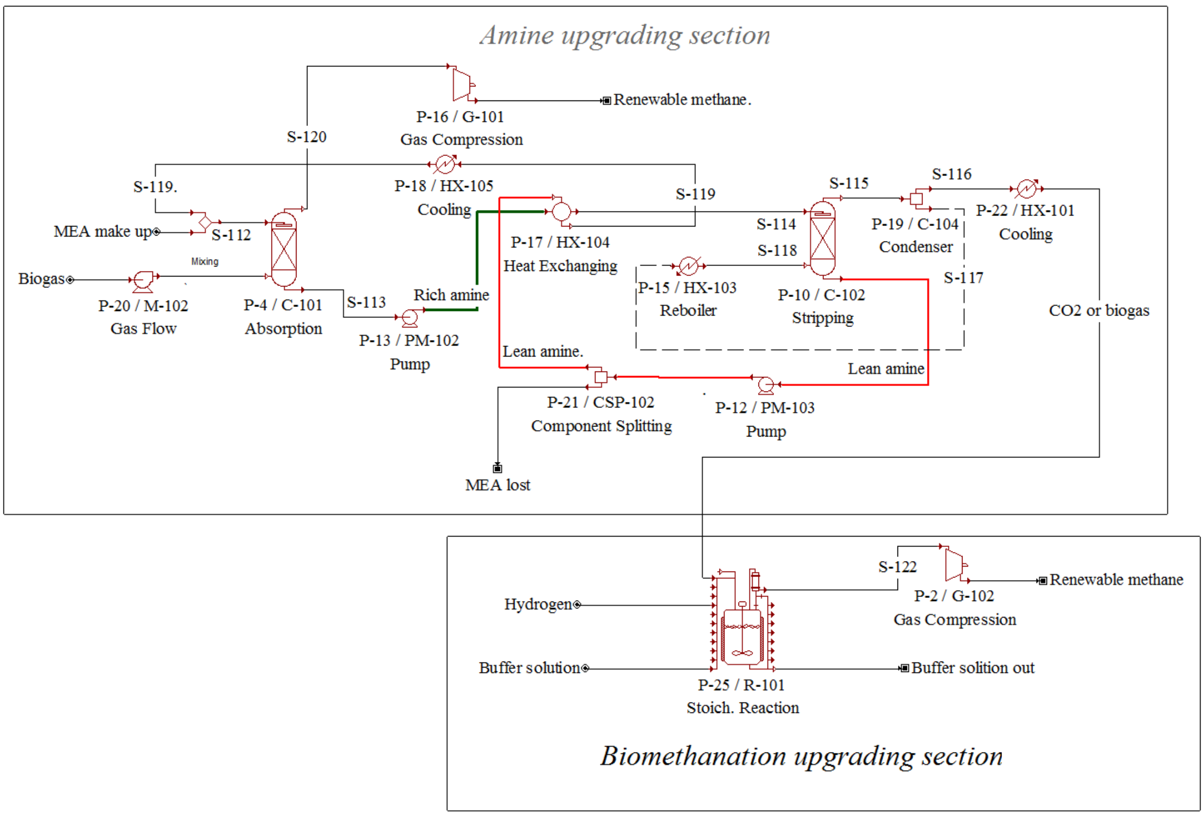


Figure 3: Upgrading section simulation

a. Solvent selection

There are many amine chemicals that dissolve into water as a solvent for biogas scrubbing such as mono-ethanol-amine (MEA), di-ethyl ethanol amine (DMEA), diethanolamine (DEA) or a mixture of methyldiethanolamine (MDEA) and piperazine (PZ)[11]. MEA ($HOC_2H_4NH_2$) was chosen for this study as previous literature has indicated it has a rapid reaction rate, low cost, ease of reclaiming, reasonable thermal stability, low molecular weight, high absorbing capacity on a mass basis, and relatively low solubility of hydrocarbons (CH_4) in the solution [37]. MEA concentrations normally range from 12 to 30%wt [38]. Park *et al*. [39] found that at the same gas – liquid ratio and re-boiler temperature, CO_2 removal efficiency is higher at 30%wt MEA solution than 20%. The disadvantage of using high amine concentrations is corrosion; however with the help of some corrosion inhibitors, 30%wt MEA solvent can be used [40]. Thus, 30%wt MEA solution is assumed in this simulation. The overall reaction of MEA with CO_2 is presented in Eq. 4 and H_2S in Eq.5 and 6.

$$2(OH-(CH_2)_2-NH_2) + CO_2 \underset{\text{High temperature}}{\overset{\text{Low temperature}}{\rightleftarrows}} (OH)-(CH_2)_2-NHCOO^- + OH-(CH_2)_2-NH_3^+ \quad \text{Eq. 4}$$

$$2(OH-(CH_2)_2-NH_2) + H_2S \underset{\text{High temperature}}{\overset{\text{Low temperature}}{\rightleftarrows}} (HOCH_2CH_2NH_3)_2S \quad \text{Eq. 5}$$

$$(HOCH_2CH_2NH_3)_2S + H_2S \underset{\text{High temperature}}{\overset{\text{Low temperature}}{\rightleftarrows}} 2HOCH_2CH_2NH_3HS \quad \text{Eq. 6}$$

The reaction between MEA and CO_2 is an exothermic reaction; for each mole of CO_2 absorbed in MEA solution, 72kJ of thermal energy is released [31]. The maximum CO_2 loading of amine solutions is 0.4 mole CO_2/mole amine.

b. Absorber and stripper designs

The absorber and stripper are packed with packing material to increase the contact time between solution and CO_2 and H_2S. The choice of packing material for the absorber and stripper is important as risk of flooding increases if surface area is excessive. Plastic palls or *Raschig* rings are usually used as packing material in absorbers but stainless steel must be applied in the stripper due to the high temperature. In addition to this, Sinnott [41] recommended that the diameter of packing size is linked to the column diameter. Plastic and metal pall rings are applied for the absorber and stripper in this study, respectively (Table 3) [41] . The absorber operates at a temperature of 40°C and pressure of 1bar, whilst the stripper operates at temperature 120°C to 150°C [31] and pressure 1.5bar to 3bar [11]. The stripper operates at 120°C and 1.5 bar in this study.

Table 3: Design data for packing [41]

Name	Size(mm)	Surface area (m^2/m^3)	Packing factor (m^{-1})
Plastic pall ring	38	128	130
Metal pall ring	16	341	230

The pressure drop of packing absorbers and strippers ranges from 15 to 50 mm of water per meter of packing height; if the liquid has foaming characteristics, the pressure drop will be halved [41]. The MEA solution is likely to foam in the absorber and stripper [42] and the loss of MEA is very small [43]; a pressure drop of 15mm water per meter of packing height (147 Pa/m) is therefore chosen in this study. The variables used in the absorber and the stripper are summarised in Table 4.

Table 4: Variables input for absorber and striper in Superpro Designer

Variables	Unit	Value	
		Absorber	Stripper
Diffusivity of CO_2 in gas phase	m^2/s	0.016	0.016
Diffusivity of CO_2 in liquid phase at 40°C	m^2/s	0.0087 [44]	0.0087 [44]
MEA 30%wt solution surface tension	N/m	0.054 [45]	
MEA 30%wt solution phase viscosity	cP	1.67 [46]	2.7 [46]
Gaseous phase viscosity	cP	0.012 [47]	0.68 [48]
Pressure drop per unit length	Pa per metre of packing height	147	147

2.2. Economic analysis

2.2.1. Capital expenditure

In this study the capital costs include for direct fixed capital (the cost paid for building a plant: equipment, installation, building, engineering, construction, etc.) and working capital (the cost needed to start the plant up and operate it to the point when income is earned). The working capital is calculated to cover 30 days of expenses for labour, raw materials, utilities (i.e., heating/cooling agents and power) and miscellaneous costs. The purchase prices of equipment to be installed are important factors in calculating the fixed capital.

The Lang factor is the ratio of the direct fixed capital cost to equipment purchase price [41], thus Lang factor will be used in this study to calculate the fixed capital of biogas plant as in the studies by Tao *et al.* [49] and Amigun *et al*. [50]. According to Tao *et al*.[49] the Lang factor of cellulosic fuels was 1.8 whilst Amigun *et al*. concluded that the Lang factor for a centralised biogas plant in Africa was 1.78 [50]. Thus, a value of 1.79 was chosen as the Lang factor for the biogas section in this study. For biogas upgrading via an amine scrubber, the Lang factor is 5 as indicated in literature [51]. The methanation reactors were given a Lang factor of 1.25 for ex-situ methanation and 1.27 if CO_2 is captured from biogas plant and injected into methanation reactor, as per Graf et al. (2014) [36]

The costs of equipment are based on: (i) built-in costs of the Superpro Designer model (dairy slurry storage tank, feedstock shredder, pump, stripper, absorber, heat exchanger, condenser and compressor); and (ii) data from literature (anaerobic digesters, gas compressors, activated carbon bed, reboiler, methanation reactor and pump – Table 5). The costs of SCADA and switch boards, tractor, pipes and biogas flare for the biogas section are calculated at 20% of the purchase equipment costs of the biogas section and all unlisted equipment (pipe, connection and amine storage tank) for the amine scrubber section is assumed at 5% of the amine equipment costs. The equipment costs of biological methanation section are taken from literature [36] and include for the compressor and any unlisted equipment. If the cost of equipment is based on a different year than the year that the plant is built, inflation or deflation is taken into account using Eq. 7 [52]:

$$C_{p,c} = C_{p,p} \left(\frac{Ic}{Ip} \right) \quad \text{Eq.7}$$

Where:

$C_{p,c}$ is the inflation-adjusted cost of equipment in current year

$C_{p,p}$ is the known cost of equipment in a previous year

Ic is the cost index for current year

Ip is the cost index for the previous year in which equipment cost is known

This research calculates the cost of product for the year 2016. The year when equipment purchased costs are used to calculate in this process is 2010 (Table 5). The chemical engineering cost indexes (CECI) of the year 2010 and 2016 are 550.8 and 535, respectively.

Furthermore, if the scale of the facility is not the same as the current scale, the cost is adjusted using Eq. 8 [52].

$$\text{New cost} = \text{original cost} \left(\frac{\text{new size}}{\text{original size}}\right)^{n} \qquad \text{Eq. 8}$$

Where:

n is the economy of scale sizing exponent (less than unity) and is dependent on the type of equipment or plant.

The base costs, original sizes and ‘n’ of process component data are presented in Table 5. For the components which ‘n’ is not known, the traditional index of 0.6 will be used [41].

Table 5. Base equipment costs and its scaling factors from literature

Component	Base cost (€)	Scale factor (n)	Base size	Sizing parameter	Base year	Ref.
Biogas plant section						
Dairy slurry storage tank	From Superpro Designer					
Feedstock shredder						
All pumps						
Silage storage pit	€503,000*					
CSTR 1	566,000	0.6	4,210	Volume (m^3)	2010	[53]
CSTR 2	180,000	0.6	1,885	Volume (m^3)	2010	[53]
Amine upgrading section						
Boiler (100kPa)	2,648	0.5	1	Mass flow rate steam (kg/h)	2010	[52]
Condenser	From Superpro designer					
Absorber						
Stripper						
Heat exchanger						
Cooler						
Methanation section						
Ex-situ methanation plant	2,464,000			5MW		[36]
Captured CO_2 from biogas plant and inject into methanation plant	2,314,700			5MW		[36]

* €900,000 for capital cost of silage storage pit [18] (if the Lang factor is 1.79, the equipment cost is €900,000/1.79 = €503,000)

2.2.2. Operating costs

Fixed and variable operating costs are the two main categories of operating costs. The fixed operating cost includes the costs of maintenance, labour and taxation. These are estimated by using factors that are normally based on direct fixed capital cost (Table 6). The personnel, laboratory and insurance costs are analysed and based on literature [52].

Table 6. Operation cost assumptions

Fixed operating costs	Cost estimation		
	Biogas plant	Methanation upgrading	Amine scrubber upgrading
Maintenance	3% equipment purchase cost [54]	3% equipment purchase cost [36]	4% direct fix capital [30]
Personnel*	2 operators	2 operators	2 operators
	1 manager		
Laboratory	15% of operating labour		
Insurance	0.7% direct fix capital		

* Annual salary for manager: €70, 000; Annual salary for operator: €40, 000

The variable operating costs consist of raw materials and utilities costs, which are calculated, based on the simulation processes in the SuperPro Designer model. The steam, cooling and chilling water costs taken from the SuperPro Designer model are €12/t, €0.05/t and €0.4/t, respectively. It is assumed that the electricity for the electrolyser to produce H_2 is produced from nearby wind farms with electricity price of €0.10/kWh, from Vo *et al.* (2017) [9] the corresponding price of H_2 is €0.147/kWh [9]. This is equivalent to €5.78/kg H_2 ((14.7c€/kWh * 3.54kWh/m^3)/0.09kg/m^3) – excluding costs of compression and storage tank. The price of grass silage feedstock is taken at €27/t and the dairy slurry is assumed free of charge, as the farmer will receive the return digestate. Since the CO_2 is taken from the biogas plant, the price of CO_2 is assumed zero. The initial amount of amine that is used for biogas upgrading is a one-off expense and not an operating cost, as this is a cost paid at the beginning of the plant operation (not annually). Therefore, the amine costs are calculated in start-up and validation expenses, the amine lost in the process is replenished every cycle.

2.2.3. Revenues

There are two products, renewable methane and digestate, which can be sold from the processes. Digestate is a fully fermented nutrient-rich material that can be utilised as an organic fertiliser, which is assumed free for farmers as they provide slurry. In order to be able to compare different pathways, the value of renewable methane from all scenarios is defined as the minimum selling prices for an 8% internal rate of return (the lowest product price that is capable of yielding a net present value of zero).

3. Results and discussion

3.1. Mass analysis

If every hour 3.5 wwt dairy slurry and 3.2 wwt grass silage are fed into the CSTR, this equates to 27,720 wwt of dairy slurry and 25,344 wwt of grass silage per year. As stated in section 2.1.1, dairy slurry and grass silage contain fixed solids (ash) (not consumed by the microorganisms in producing CH_4), nutrients and VS (which are used mostly as energy for microorganisms to produce CH_4). Total annual renewable methane production from scenario 1, 2 and 3 are: 3.44 Mm^3; 6.63 Mm^3 and 6.64 Mm^3, respectively. It is noted that the renewable methane is not only pure methane, it contains trace H_2S and small amounts of CO_2 that could not totally be eliminated. The mass flows in and out of the three scenarios are summarised in Figure 4. If 3.28Mm^3 pure methane is generated per year and the total VS added per year is 8,953t , then one tonne of VS can be calculated to produce 366m^3 pure methane; this is consistent with yields suggested in previously literature [55].

The digestate composition was made up of fixed solids, nitrogen, phosphorus, potassium, water and some VS from slurry and grass, which was not consumed by the microorganisms. In total, 44,539 t digestate are discharged per year. Among the three scenarios, only CO_2 out from scenario 1 is emitted directly to the atmosphere because it is not re-used in a further step (as in scenario 2 and 3). The higher CO_2 content in renewable methane suggests that that energy content in renewable methane from scenario 2 (95.5%vol. of CH_4) is lower than that of scenario 3 (97% vol. of CH_4). The quantities of methane produced are almost double in scenario 2 and 3 as compared to that of scenario 1 (as biomethane converts each volume of CO_2 to an equivalent volume of CH_4). Due to the small amount of CO_2 in renewable methane, which results from biogas upgrading, the total quantity of CO_2 from the amine scrubber is less than the total CO_2 volume in the biogas which is fed directly to the ex-situ biological methanation reactor. H_2 is assumed to be fed into the methanation reactor at the recommended ratio of 4 H_2: 1 CO_2. The amount of H_2 fed into the methanation reactor in scenario 2 is again less than that of scenario 3. In order to ensure less than 2%vol. of H_2 in the final gas as required for gaseous fuel [56], 99% of H_2 which is fed into methanation reactor needs to be converted to CH_4. A quantity of 3,472 kg mono-ethanol-amine, which is fed at the start of the process, is recycled. However, annually 205 kg mono-ethanol-amine is lost due to evaporation, thus this amount needs to be replenished to the process to compensate for this loss.

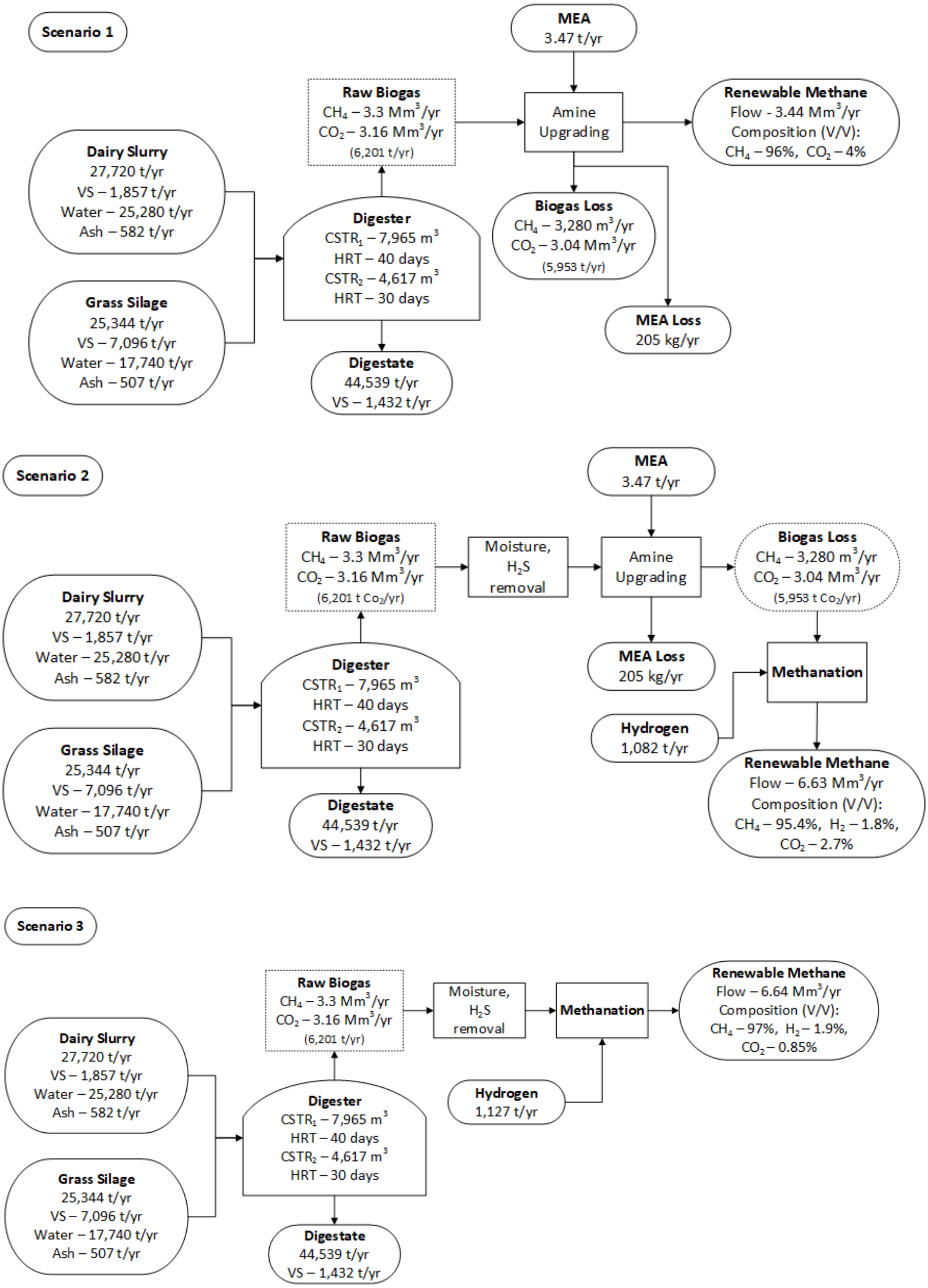


Figure 4: Mass balance of three scenarios

3.2. Energy analysis

The different types of energy, which are used in the three scenarios, are: electricity; steam; cooling and chilling water. Scenario 2 contains three sections (biogas, upgrading by amine and methanation) of the process; the energy consumption (electricity and steam (heat)) in this scenario are analysed (Figure 5) to understand the energy consumption in each section. Upgrading biogas by an amine scrubber only requires a very small amount of electricity (0.023kWh/m^3 renewable methane) but has the highest energy requirement for heating. This is because a large amount of heat is required to regenerate CO_2 from rich amine solution. The biogas section has a lower consumption of standard power than that of methanation due to electricity requirement for mixing H_2 into solution. The total energy consumed in the biogas section is lower than from the amine scrubber section.

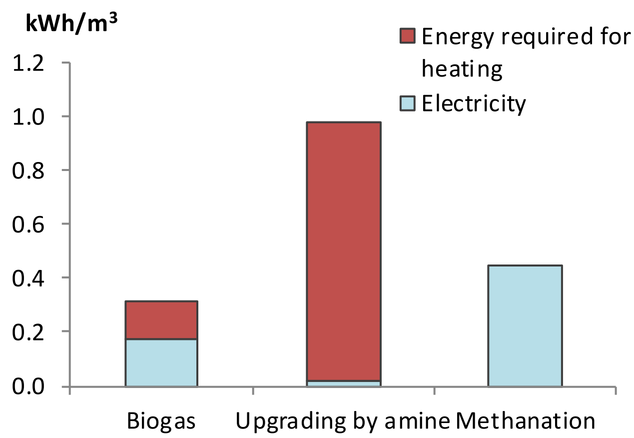


Figure 5: Energy consumptions per m^3 of renewable methane of different sections in Scenario 2

By dividing each energy subdivision for a whole year by the total renewable methane produced, the energy consumed per m^3 of renewable methane can be calculated as shown in Figure 6. The electricity requirement is higher for scenarios 2 and 3 than for scenario 1. However, the amount of steam used in scenario 3 is lowest as there is no amine scrubber.

Although the amine scrubber for both scenarios 1 and 2 consumes steam, it should be noted that the renewable methane production from scenario 2 is almost double that of scenario 1. Thus, for one m^3 of renewable methane, the steam used for scenario 2 is practically half that of scenario 1. Total energy consumption is highest in scenario 1 and lowest in scenario 3. If one m^3 of renewable methane contains 10kWh, the energy consumption for scenario 1, 2 and 3 account for 25.7%; 18.7% and 9% respectively of final produced energy.

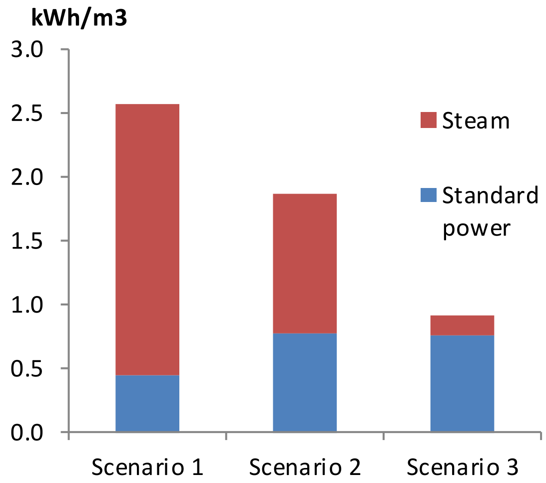


Figure 6: Energy consumptions per m^3 renewable methane of different scenarios

3.3. Economic analysis

3.3.1. Direct fixed capital cost

The biogas section accounts for the highest direct fixed capital investment cost (€3.91M), followed by the cost of upgrading by methanation (€2.9 -3.1M) and subsequently the cost of upgrading by amine (€1.57 M). The major equipment costs and their sizes for scenario 2 and 3 can be found in the Appendix B, C & D. Table 7 gives an example of equipment purchase costs for scenario 1. The name of sections can be seen in Figures 2 and 3. In this study, hydrogen was purchased separately; the analysis did not include the cost of the electrolysis plant.

Table 7. Component purchase costs of scenario 1

Name	Description	Cost (€)
1. Biogas section		
SL-101	Grass silage storage pit For storing 30,000 t grass silage/year	503,000
V-101	Slurry Receiver Tank Vessel Volume = 170 m^3 (2days storage)	70,000
SR-101	Shredder Rated through put = 3,200kg/h	56,000
PM-101	Centrifugal Pump Pump Power = 0.51 kW (pump and mixer)	7,000
AD-101	Anaerobic Digester Vessel Volume = 7965 m^3	805,000
AD-102	Anaerobic Digester Vessel Volume = 4616 m^3	308,000
Total 1		**1,749,000**
2. Amine scrubber section		
M-102	Blower	3,000
C-101	Absorber Flow rate of biogas 813m^3/h; column diameter 1m; column height 15m.	35,000
C-102	Stripper Flow rate 403 m^3/h; column diameter 1m; column height 15m.	35,000
PM-102	Centrifugal Pump Pump Power = 0.69 kW	10,000
HX-104	Heat Exchanger Heat Exchange Area = 15.63 m^2	12,000
HX-105	Heat Exchanger Heat Exchange Area = 9.69 m^2	8,000
PM-103	Centrifugal Pump Pump Power = 0.64 kW	12,000
HX-103	Reboiler	84,000
C-104	Condenser	22,000
G-101	Centrifugal Compressor Compressor Power = 30.13 kW	47,000
Total 2		**268,000**
Unlisted Equipment		**451,000**
Total 1+2 + unlisted equipment		**2,468,000**

3.3.2. Economic comparison of overall process.

For an NPV equivalent of zero, an electricity price to produce H_2 of €0.10/kWh and a grass silage production cost of €27/t, the minimum selling price of one m^3 renewable methane (CH_4) excluding tax for scenario 1, 2 and 3 are €0.76; €1.50 and €1.43, respectively. A detailed economic breakdown of the three scenarios is presented in Table 8. Total capital investment cost is highest in scenario 2, because this scenario includes the biogas plant, biogas upgrading and biomethanation. The capital investment of scenario 3 is slightly higher than that of scenario 1 despite a more unique upgrading method. As mentioned in section 2.1.1, the removal of H_2S below 1-ppm costs €0.01/m^3 biogas, which means it costs €0.01/m^3 renewable methane in scenario 2 and 3 as the final volume of renewable methane in those scenarios is similar to the volume of biogas. The costs are €0.02/m^3 renewable methane in scenario 1 as the volume of renewable methane after eliminating CO_2 is only half of biogas volume.

Table 8: Overall economics of three scenarios

	Scenario 1	Scenario 2	Scenario 3
Total capital investment (M€)	5.486	8.81	7.8
Operating cost (M€/yr)	1.97	8.98	8.6
Renewable methane revenue (M€/yr)	2.5	9.9	9.28
Produced renewable methane (Mm^3/year)	3.44	6.636	6.642
Minimum selling price (€/m^3)	0.76	1.50	1.43
Net unit production cost (€/m^3)	0.57	1.35	1.3

The costs of raw materials account for the largest percentage of the operating costs in scenario 2 and 3 and is slightly lower than utilities cost in scenario 1 (Figure 7). It may be noted that the facility-dependent costs consist of maintenance and miscellaneous costs. The cost of grass silage accounts for 38% in scenario 1 and approximately 12% in scenarios 2 and 3. The cost of CH_4 may be reduced if feedstocks are available free of charge such as for food waste (which may even accrue a gate fee) or slaughterhouse waste [9]. Because of high H_2 costs, the raw material cost in scenario 2 is 77% and 84% in scenario 3. Approximately 90% of the H_2 cost is for the purchase of H_2.

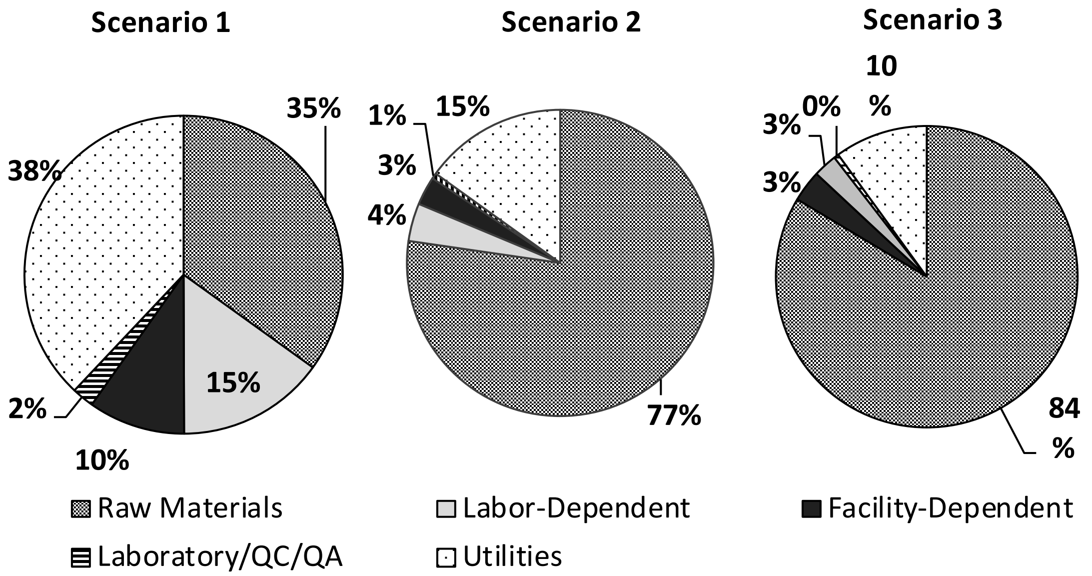


Figure 7. Operation costs in percentage of three scenarios

3. 4. Comparison of data with literature

When comparing the CAPEX per m^3 methane per year to previous studies analysing different feedstocks, the CAPEX of the plants in the three scenarios fit within the range of previous research (Figure 8).

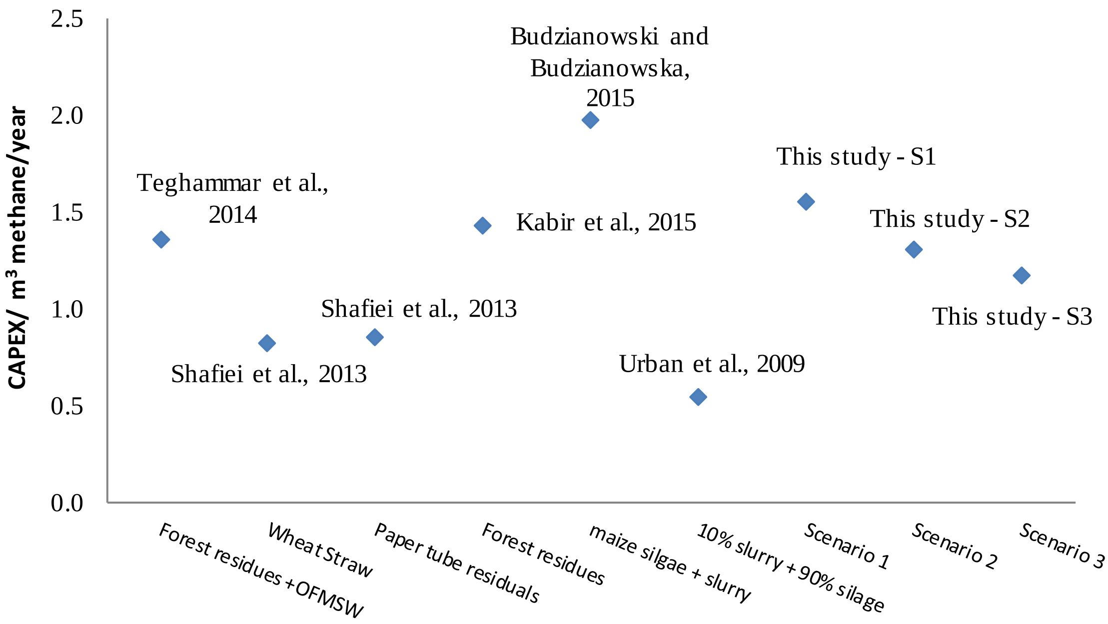
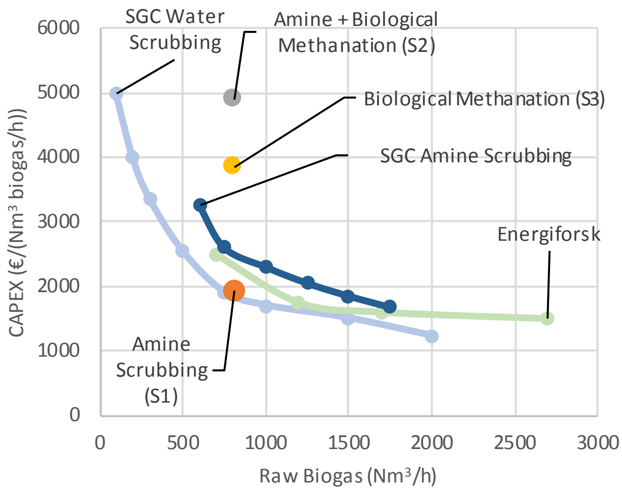


Figure 8: CAPEX/ m^3 methane/ year in this study and previous researches

Note: Teghammar et al. [57], Shafiei et al. [58], Urban et al. [59], Kabir et al. [60], Budzianowski and Budzianowska [61]

The CAPEX of amine scrubbing was compared to other biogas upgrading methods (Figure 9). The CAPEX of the amine scrubber from this study (€1,936/Nm^3/h) was close to values reported in the literature. Although water scrubber upgrading is the most popular method, it is not ideally suited to carbon capture and reuse as it does not produce a free stream of CO_2. In addition to this, the losses of methane are relatively high for water scrubbing in comparison to amine scrubbing [62].



S1: Scenario 1 Amine scrubbing

S2: Amine scrubbing and biological methanation

S3: Biological methanation

Figure 9: Comparison of CAPEX of different upgrading methods[63, 64].

The energy demand of amine upgrading in this study was compared with the data in the literature. Similar power consumption was reported in this study as with literature [11], while the heat demand to release CO_2 from amine solution in this research was approximately three times higher than that reported in literature (Figure 10). The rationale behind the higher heat requirement was based on the CO_2 content in the biogas. To release 1 ton of CO_2, 4GJ of heat was required [29, 30, 32]; the total CO_2 produced in scenario 1 was 5,953 tons. The total heat required to release all the CO_2 was 23,812 GJ. When the heat required was recalculated based on the methane-produced, this generated a figure of 1.9 kWh/Nm^3 methane. This study reported a heat requirement of 1.85 kWh/m^3 methane from calculations in SuperPro.

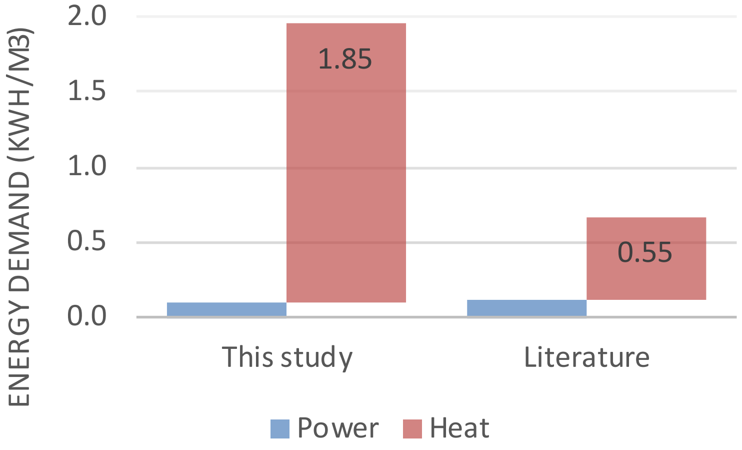


Figure 10: Comparison of energy demand of amine upgrading with literature data [11].

3.5. Sensitivity analysis

As indicated, raw material costs contribute to a high percentage of the operating costs for the three scenarios. However, the production price of grass silage and the cost of H_2 are variable. The cost of H_2 production varies in that the cost of electricity fluctuates with temporal supply and demand issues. For example, at times of excess production electrolysis is seen to have a stability function in the electricity grid and the cost of the electricity may be minimal, reducing the price of H_2. A number of factors influence the cost of grass silage, including: the number of harvests; fertiliser application; fuel usage; supply and demand. Thus, a sensitivity analysis will take into account the variation in grass silage cost per tonne at prices of €13.5, €27, €40.5 and electricity cost per kWh at €0.05; €0.10 and €0.15.

If grass silage costs increase by 50%, it is calculated that the cost of renewable methane will increase by €0.10 per m^3 in scenario 1. The renewable methane cost only increases by €0.05 per m^3 for scenarios 2 and 3. This due to the fact that approximately half of the renewable methane is produced by combining H_2 and CO_2, thus, the grass silage cost does not have as much of an impact (Table 9).

Table 9: Effect of grass silage price on renewable methane production

Grass silage price per t	Renewable methane price at electricity cost €0.10/kWh		
	Scenario 1	Scenario 2	Scenario 3
€13.5	€0.66	€1.45	€1.38
€27	€0.76	€1.50	€1.43
€40.5	€0.86	€1.55	€1.48

Hydrogen cost will not affect the renewable methane cost in scenario 1 as this process does not include for biomethanation. If the electricity price changes by €0.05 per kWh (by 50%), the renewable methane cost will change by approximately €0.41 for scenario 2 and €0.43 for scenario 3 (Table 10). Therefore, the electricity price used to calculate H_2 costs has a very significant effect on the price of renewable methane.

Table 10. Effect of electricity price (to produce hydrogen) on renewable methane production

Electricity price change per kWh	Renewable methane price at grass silage €27 per t		
	Scenario 1	Scenario 2	Scenario 3
€0.05	€0.76	€1.09	€1.00
€0.10	€0.76	€1.50	€1.43
€0.15	€0.76	€1.91	€1.85

Besides grass silage cost and electricity price, the authors were aware that the quantity of renewable methane could be increased if higher quantities of grass silage feedstock were fed to the digester. Currently the ratio modelled was 80% VS from grass silage and 20% VS from dairy slurry. Therefore, the effect of increasing the percentage VS from grass silage was also analysed. A maximum grass silage scenario was investigated with 45,000wwt of grass silage and a minimum dairy slurry concentration of 5,238 t wwt (for nutrient addition) assumed to be fed to the digester annually. The cost per m^3 renewable methane calculated for scenarios 1, 2 and 3 at €27/t grass silage and €0.10/kWh electricity (to produce H_2) was €0.74; €1.45 and €1.40, respectively. This equates to a €0.02; 0.05 and 0.03 drop in price for scenario 1, 2 and 3, respectively.

4. Conclusion

The process simulation performed using SuperPro Designer has shown that among three scenarios, the production cost of renewable methane from a grass and slurry fed biogas plant using traditional upgrading (amine scrubber in this study) was the cheapest (€0.76/m^3 renewable methane). If surplus renewable electricity needs to be stored as methane, the direct methanation of biogas is more economically advantageous than capturing the CO_2 from upgrading and subsequently providing methanation of the captured CO_2. From an environmental/sustainability/decarbonisation perspective, the renewable methane generated from biological methanation (Scenario 3) is more sustainable than amine scrubbing (Scenario 1) as no CO_2 is released to the atmosphere. The key drivers for feasible processes include technical advancements such as a single-unit upgrading systems with carbon capture, increases in the efficiency of biomethanation, reduced costs for feedstock, reduced costs for electricity in the production of hydrogen and reduced electrolyser CAPEX. Power to gas systems are relatively new technologies that still need maturity to allow reduction in investment costs.

Acknowledgements

This research is funded by the Science Foundation Ireland (SFI) centre MaREI (12/RC/2302) with industrial co-funding from ERVIA, Gas Networks Ireland (GNI) through the Gas Innovation Group.

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