Sveriges lantbruksuniversitet
Swedish University of Agricultural Sciences

Institutionen för energi och teknik

System concept for small-scale biological methanation using electrolysis and trickle bed reactor

- with the aim of upgrading biogas to vehicle fuel quality

Konceptsystem för småskalig biologisk metanisering med elektrolys och trickle bed reaktor

- *med mål att uppgradera biogas till fordonsbränsle*

Gustav Larsson

Civilingenjörsprogrammet i energisystem

Examensarbete 2021:14
ISSN 1654-9392
Uppsala 2021

System concept for small-scale biological methanation using electrolysis and trickle bed reactor - with the aim of upgrading biogas to vehicle fuel quality

Konceptsystem för småskalig biologisk metanisering med elektrolys och trickle bed reaktor – med mål att uppgradera biogas till fordonsbränsle

Gustav Larsson

Handledare: Johan Andersson, RISE
Ämnesgranskare: Leandro Janke, institutionen för energi och teknik, SLU
Examinator: Åke Nordberg, institutionen för energi och teknik, SLU

Omfattning: 30 hp
Nivå, fördjupning och ämne: Avancerad nivå, A2E, teknik
Kurstitel: Examensarbete i energisystem
Kurskod: EX0724
Program/utbildning: Civilingenjörsprogrammet i energisystem 300 hp
Kurskoordinerande institution: Institutionen för energi och teknik

Utgivningsort: Uppsala
Utgivningsår: 2021
Serietitel: Examensarbete (Institutionen för energi och teknik, SLU)
Delnummer i serien: 2021:14
ISSN: 1654-9392
Elektronisk publicering: http://stud.epsilon.slu.se

Nyckelord: biological methanation, biogas upgrade, Methane, small-scale methanation

Sveriges lantbruksuniversitet
Swedish University of Agricultural Sciences

Fakulteten för naturresurser och jordbruksvetenskap
Institutionen för energi och teknik

Publicering och arkivering

Godkända självständiga arbeten (examensarbeten) vid SLU publiceras elektroniskt. Som student äger du upphovsrätten till ditt arbete och behöver godkänna publiceringen. Om du kryssar i **JA**, så kommer fulltexten (pdf-filen) och metadata bli synliga och sökbara på internet. Om du kryssar i **NEJ**, kommer endast metadata och sammanfattning bli synliga och sökbara. Fulltexten kommer dock i samband med att dokumentet laddas upp arkiveras digitalt.

Om ni är fler än en person som skrivit arbetet så gäller krysset för alla författare, ni behöver alltså vara överens. Läs om SLU:s publiceringsavtal här: https://www.slu.se/site/bibliotek/publicera-och-analysera/registrera-och-publicera/avtal-for-publicering/.

☒ JA, jag/vi ger härmed min/vår tillåtelse till att föreliggande arbete publiceras enligt SLU:s avtal om överlåtelse av rätt att publicera verk.

☐ NEJ, jag/vi ger inte min/vår tillåtelse att publicera fulltexten av föreliggande arbete. Arbetet laddas dock upp för arkivering och metadata och sammanfattning blir synliga och sökbara.

Abstract

In the present study, the concept of a biological H_2 methanation (BHM) system was created for four cases of scale which are determined by electrolyser scale. The system design is aimed to upgrade existing biogas to vehicle fuel quality, with a concentration of CH4 above 95% and H2S removal. The cases of scale and type of electrolyser are: 4.8 kW AEM electrolyser, 20 kW AEL electrolyser, 100 kWe AEL electrolyser and 550 kWe PEM electrolyser. Each case of scale can upgrade a biogas flow of 0.7, 2.4, 14.5 and 73.9 Nm^3 respectively. A trickle bed reactor design at thermophilic conditions was chosen for the systems methanation process. A MATLAB model was created to simulate energy- and mass flows for the system. The simulation also includes economic parameters such as OPEX and CAPEX. Results of the simulation are presented as levelized cost of CH4 production (€/kWh) and specific CAPEX (€/kW_e). Simulations of the system show a high upgrading performance with an output gas of above 95% CH4 with H2S removal. The system also increases CH_4 yield of 60%. The system performs comparatively to traditional upgrading method. The economic results show that the system has an upgrading cost of 0.37 to 0.089 €/kWh and specific CAPEX of 3830 to 22 500 €/kW. The system cannot be considered economically competitive to traditional upgrading when no additional cost reductions are applied. The concept of upgrading by BHM also reduces carbon emissions from biogas production giving the system a good chance of receiving subsidies from greenhouse gas reduction initiatives. Larger scales of the system can then reach competitive upgrading costs by utilizing subsidies, electricity price reductions and heat recovery.

Keywords: Biological methanation, Biogas upgrade, Methane, Small-scale methanation

Populärvetenskaplig sammanfattning

Vid ett cirkulärt system kan biogasproduktion anses vara viktig. Genom att producera biogas från avloppsslam, matavfall eller annat organiskt avfall kan energi utvinnas samt gödslingsmedel produceras. Biogasen som produceras innehåller en hög halt koldioxid (35–40%). Gasen kan uppgraderas genom att avlägsna koldioxiden och 2019 producerades 2.1 TWh biogas varav 64 procent av all biogas uppgraderas för att användas till fordonsgas. Däremot är uppgradering av biogas dyr och är oftast reserverad för större anläggningar. För mindre anläggningar som gårdsanläggningar eller mindre reningsverk uppgraderas sällan biogasen och i stället används rågas för värme- och elproduktion. Det finns då ett behov att hitta nya kostnadseffektiva metoder för uppgradering.

Biologisk metanisering är en process där mikroorganismer omvandlar koldioxid och vätgas till metan och vatten. Processen är en del av den mikrobiologiska processkedjan för att producera biogas. Metaniseringsprocessen kan då användas till att öka halten och mängden metan ur en biogasprocess genom att injicera vätgas. Detta kan göras direkt i rötkammare eller i ett externt kärl med mikroorganismer där koldioxid/biogas tillförs. Eftersom metoden kräver vätgas produceras oftast vätgasen genom elektrolys, som omvandlar vatten till vätgas när en elektrisk spänning tillförs.

I detta examensarbete utformades ett konceptuellt småskaligt uppgraderingssystem som med hjälp av biologisk metanisering och elektrolys kan uppgradera biogas till fordonsgas. Metaniseringen utförs i en ”trickle bed” reaktor där mikroorganismerna är immobiliserade i en fast bädd där sedan CO_2 och H_2 injiceras i botten. Systemet konstruerades för fyra olika skalor och mättes på installerad kapacitet för elektrolysen: 4.8 kW, 20 kW, 100 kW och 550 kW. Dessa skalor kan uppgradera ett biogasflöde på 0.7, 2.4, 14.5 respektive 73.9 Nm^3 biogas. En modell för systemets skalor skapades. Energibalans, massbalans och metanproduktion simulerades. En ekonomisk analys utfördes även med mål att jämföra uppgraderingskostnaderna gentemot traditionell uppgradering som används i Sverige. Beräkningarna baseras på termofila förhållanden (55°C) för metaniseringsprocessen.

Resultaten visade att systemet har potential till att användas för uppgradering. Systemet når en producerad gaskvalité för fordonsgas. Kostnader för systemet visade sig vara starkt beroende av elektrolys- och elkostnader och detta fenomen förstärktes vid högre skalor. Uppgraderingskostnader för systemet nådde 0.4 €/kWh till 0.09 €/kWh. Kostnaderna för bassimuleringarna kan inte anses vara tillräckligt låga för att motivera en användning av systemet. Däremot kan högre skalor (100 kW till 550 kW elektrolyskapacitet) nå en viss ekonomisk konkurrenskraft med hjälp av bidrag från initiativ med mål att sänka växthusgasutsläpp.

Executive summary

The present study´s purpose was to create a concept of a biological methanation system with the purpose of upgrading biogas at a small scale. The system was designed at 4 cases of scale based on electrolyser capacity: 4.8 kWe, 20 kWe, 100 kWe and 550 kWe which correlates to a biogas flow of 0.7, 2.4, 14.5 and 73.9 Nm^3 respectively. The system was designed to use electrolysis and biological methanation through a trickle bed reactor at thermophilic conditions (55 °C). Mass flow, energy balance, production rate, specific CAPEX and levelized cost of CH_4 was investigated through simulations in a MATLAB model designed for the system.

Results indicate that the system can upgrade biogas to vehicle fuel quality and increases the yield of CH_4 by 60% for raw biogas of 40%-vol CO_2. The system cannot be considered economically competitive at any scale. Cost reductions such as subsidies, electricity price reductions or heat recovery cases at 100 and 550 kW can be competitive. Future developments in electrolysis and biological methanation technology are expected to reduce production costs.

Acknowledgements

This master thesis was performed for master’s Programme in Energy systems Engineering at Uppsala university and SLU. The project was made in cooperation with Research institutes of Sweden (RISE) and Swedish University of Agricultural Sciences (SLU).

I would like to thank my supervisor at RISE, Johan Andersson who have provided me, with his expertise, guidance and help through the whole project. I would also like to thank everyone else at the “Kretslopp”-department of RISE who have welcomed me to gatherings, meetings and given me presentation opportunities.

I would also like to give my thanks to my subject reviewer, Leandro Janke for his reflections and feedback that improved the quality of the project.

Finally, I would like to thank my family and partner who has shown me unending support both through the thesis project and years of education.

Gustav Larsson
Uppsala 2021

Table of contents

List of tables

List of figures

Abbreviations

HLR	Hydrogen loading rate
CHP	Combined heat and power
TBR	Trickle bed reactor
kWe	kW electrolyser capacity
BHM	Biological hydrogen methanation
PEM	Proton exchange membrane
AEL	Alkaline electrolyser
AEM	Anion exchange membrane
MER	Methane evolution rate
CO_2	Carbon dioxide
CH_4	Methane
H_2	Hydrogen gas
CSTR	Continuous stirred tank reactor
SOEC	Solid oxide electrolyser
LCOCH4	Levelized cost of methane production
COCH4	Specific investment cost of methane
CAPEX	Capital expenditures
OPEX	Operating expenditures
FLH	Full load hours
LHV	Lower heating value
HHV	Higher heating value

1. Introduction

Biogas is an important part of today's Swedish energy sector. The scale of biogas plants varies from small-scale such as plants located at farms to large-scale industrial size plants. Biogas can be used directly as a source of electricity and/or heat generation, but a more common option is to upgrade the biogas to high concentrations of methane (>95%) to be used as vehicle fuel. In 2019 64% of all biogas produced in Sweden is upgraded to be used as fuel or other applications that require a high concentration of methane (CH_4) (Energigas Sverige 2019). Upgrading is however reserved mostly for larger biogas plants due to the high costs of upgrading equipment. Few small-scale plants such as farm-based biogas plants or municipal wastewater treatment plants use upgrading and most usage of small-scale produced biogas is used for heat and electricity which is a less lucrative option (Eliasson 2015). This creates interest in new technology and methods of upgrading biogas that can fit a small-scale system.

Methanation is a process of converting CO_2 to CH_4 and is a promising method for upgrading biogas. With traditional upgrading, the CO_2 will be removed from the biogas to increase the concentration of CH_4. The removed CO_2 becomes a by-product and is vented. Methanation however uses H_2 to convert the excess CO_2 to CH_4, effectively raising the concentration and CH_4 content of the biogas. Using this method in a biogas production process can potentially increase both the yield and quality of the biogas produced and can supplement or replace traditional upgrading (Lecker et al. 2017). The methanation process can either be biological or catalytic but serve the same function. While catalytic methanation has been proposed more often in large-scale applications, biological methanation could be more suitable for upgrading biogas from small-scale plants due to its higher tolerance for gas impurities (Strübing et 2017).

1.1. Purpose

The project aims to develop concepts for small-scale upgrading of biogas based on biological methanation at four different scales. The systems are aimed to produce biogas of vehicle fuel quality, meaning a concentration of 95% CH4 or higher with an electrolyser capacity of 4.8 kW, 20 kW, 100 kW and 550 kW. The purpose of this is to evaluate the performance and economy of available technologies of biological methanation and H_2 generation at lower scales and conceptualize a system. The following goals for the project are:

- Describe a system at four scales that fulfils the aim of producing output gas of CH_4 concentration up to 95%.
- Simulate energy balance, mass balance and CH_4 production of the system.
- For each scale, calculate the specific investment cost (€/kW_{CH4}) and the Levelized cost of CH_4 production from methanation (€/kWh_{CH4})
- Analyse the effects of CAPEX, OPEX, and operational hours to investigate the effects on economic performance.

2. Background

2.1. Small-scale biogas production in Sweden

Biogas is produced through the anaerobic digestion of organic material in a unit called digester, which can vary in size and type. The organic material also varies from crops, slaughter residues to organic waste. Biogas directly produced from the digester has a CH_4 concentration of 60-75% where the rest is mostly CO_2 (SGC 2012). The CO_2 is a by-product of the process, and some biogas plants use upgrading methods to remove the by-product. In 2019, 64% of all biogas produced in Sweden was upgraded to be used as fuel or for other energy systems that require high concentration of CH4 (Energigas Sverige 2019). If the biogas will be used as fuel, some conditions must be met. The biogas needs to have a CH_4 concentration above 95% and have a concentration of hydrogen sulfide (H_2S) of 23 ppm or lower (SGC 2012).

Upgrading biogas is expensive and is not as commonly used in small-scale biogas plants. Farm-based biogas plants usually use raw biogas for combined heat and power generation (CHP). In a study from 2015, 31 farm-based biogas plants were evaluated where 27 were not upgrading biogas. 24 of the farms are using the biogas for CHP. The report also shows that 29% of all energy produced are not used or sold, resulting in large losses (Eliasson 2015).

2.2. Biological methanation

The biogas production process can take place in an anaerobic environment at either mesophilic temperature conditions (37 °C) or thermophilic (55-65 °C). The pH requirements of the process are 6.2-8.5 (Rusmanis et al. 2019). The full process is a chain of biological processes but for the present study’s purpose, the two most important processes are covered. These are hydrogenotrophic methanogenesis or so-called biological hydrogen methanation, and acetoclastic methanogenesis which both takes place at the very end of the chain. The former is done by hydrogenotrophic methanogens, which is a type of archaea. With the use CO_2 and H_2 the methanogens form CH_4 and H_2O which is described in formula 1 (Rusmanis et al. 2019).

$$4H_2 + CO_2 \rightarrow CH_4 + 2H_2O \qquad -167\ kJ\ \Delta G° \qquad \text{(Formula 1)}$$

The latter is done by acetoclastic methanogens which converts acetate to CH_4 and CO_2 following the formula (Adnad 2019):

$$CH_3COOH \rightarrow CH_4 + CO_2 \qquad -31\ kJ\ \Delta G° \qquad \text{(Formula 2)}$$

Both processes are a crucial last step of biogas production process. Biological H_2 methanation (BHM) works as an extension to the biogas production process. BHM purpose is to use biological methanation to convert CO_2 to CH_4. In BHM, H_2 is injected into the biogas process to create higher conversion rates from the methanogens. The H_2 can be directly injected into the digestion chamber, in which the BHM will take place. This method is called "in-situ" methanation. Alternatively, the H_2 can be injected together with biogas or CO_2 from the digestion chamber into a separate "methanation reactor". In this reactor hydrogenotrophic methanogenesis is done by colonies of methanogens. This method is called "ex-situ" methanation. Both methods increase the concentration and yield of CH_4 of the output gas. This means that BHM can be an alternative for upgrading biogas (Rusmanis et al. 2019).

2.2.1. In-situ BHM

Figure 1 shows the concept of in-situ methanation. H_2 produced from an electrolyser is injected directly into a digestion chamber, in which common anaerobic digestion is taking place. To maximize the contact between methanogens and H_2 the gas is injected through mixing or by diffusion (Rusmanis et al. 2019). While yielding higher production of CH_4, in-situ methanation has a limiting factor, which is the partial pressure of H_2 (Lecker et al. 2017).

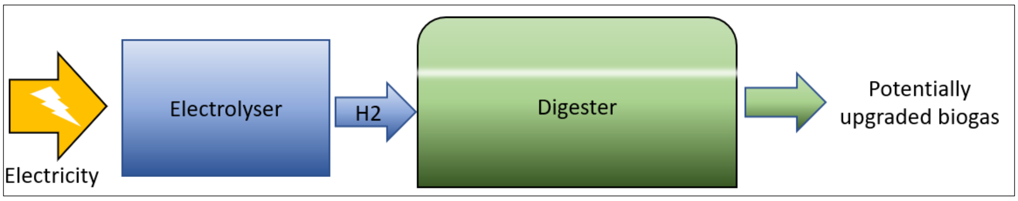

Figure 1: Concept of in-situ methanation. H_2 that is produced by an electrolyser is injected into a digester chamber, increasing the H_2 gas partial pressure.

The partial pressure of H_2 and the inhibition of acetogens

When applying in-situ BHM it is important to control the injection of H_2 carefully. Raising the concentration of H_2 will lead to an increase of hydrogenotrophic methanogens, which in turn increases the conversion rate of CO_2. However, higher concentrations of H_2 will inhibit another microorganism group called syntrophic acetogens. These microorganisms are responsible for the formation of acetate, which is needed for acetoclastic methanogenesis described in formula 2. As previously mentioned, this process is a crucial part of the biogas production process (Adnad 2019; Benjaminsson et al. 2013). If the inhibition of the syntrophic acetogens rises too high, the biogas process suffers inefficiencies and the production rate of CH_4 will be lowered. Therefore, constant monitoring of H_2 concentration inside the digester is necessary. This results in the in-situ BHM having a limitation on how much H_2 can be injected, thus, resulting in limiting CH_4 production (Lecker et al. 2017).

2.2.2. Ex-situ BHM

An ex-situ BHM process is shown in figure 2. By using an external reactor after the digestion chamber, the environment can be optimized for hydrogenotrophic methanogens in said reactor. This can be achieved without causing inhibition to the biogas process which takes place in the digestion chamber. Ex-situ processes are generally more effective than in-situ due to the higher concentrations of H_2 that can be applied to the process (Lecker et al. 2017). Biogas or pure CO_2 is injected into the reactor for conversion. Due to the stoichiometric conditions of formula 1, a ratio between H_2 and CO_2 of 4:1 needs to be maintained for full CO_2 conversion. A fraction of the carbon of the injected CO_2 will be transformed to biomass and practical experiments have shown a ratio of 3.76:1 is optimal (Burkhardt & Busch 2015).

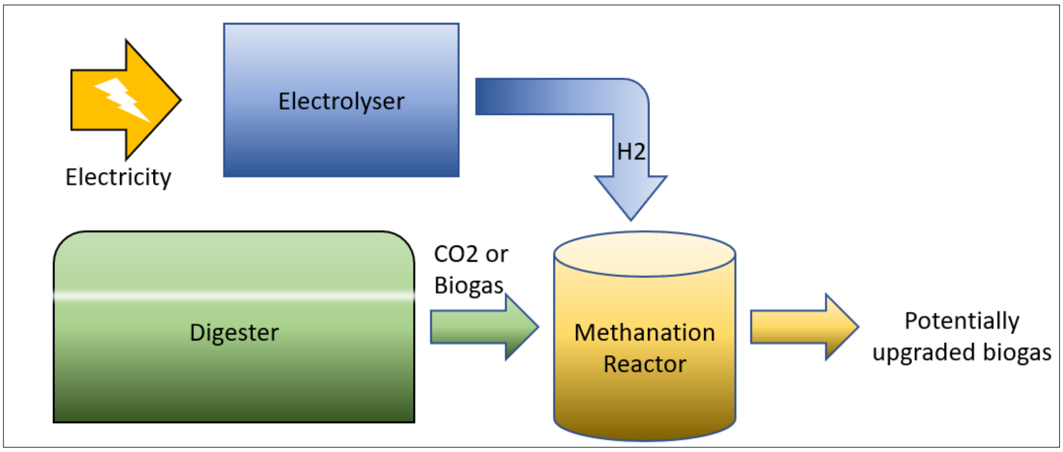

Figure 2: Concept of ex-situ methanation. H_2 *produced by an electrolyser is injected together with biogas/*CO_2 *into an external methanation reactor where BHM take place. Output gas from the external reactor has a higher yield of* CH_4*.*

Solubility of H_2

The efficiency of the BHM process increase by maximizing contact between methanogens and H_2. To do this, H_2 needs to be solved in the process liquid. H_2 gas has lower solubility in liquid than CO_2. H_2 solubility increases with lower temperatures but the methanogens requirement of mesophilic or thermophilic makes lowering the temperature impossible. This makes the solubility of H_2 a limiting factor in BHM reactors. Therefore, the reactors need to have a high gas-liquid mass transfer, meaning that the H_2 can diffuse into the liquid medium. The ability to diffuse the H_2 gas for a reactor can be described as the reactors H_2 volumetric gas-liquid mass transfer coefficient (k_La). The k_La can be increased by different means, such as mixing of process liquid, diffusion through ceramic material, H_2 recirculation or other methods for maximizing contact area between gas and liquid. Reactors using mechanical mixing such as continuous stirred tank reactor (CSTR) increase its k_La through mixing, while trickle bed reactors (TBR) increase the surface area of gas and liquid by trickling the liquid through a fixed bed and injecting gas at the bottom of the reactor (Rusmanis et al. 2019).

2.2.3. Methanation reactors

There are many different types of methanation reactors, both in-situ and ex-situ. The report from Rusmanis et al. (2019) “Biological hydrogen methanation systems – an overview of design and efficiency” summarizes different types of reactors and their performances. Due to the purpose of this thesis, only reactors that are reported to reach concentrations above 95% in the summary by Rusmanis et al. (2019) will be described. Except for CH_4 concentration other important performance measurements are H_2 loading rate (HLR) and CH_4 evolution rate (MER). HLR measures the volume of H_2 which is injected into the reactor per total reactor volume and day ($V_{H2}/V_{reactor}$ * day). MER measures CH_4 production of the methanation process ($V_{CH4}/V_{reactor}$ * day). It can also be noted that output gas from reactors needs postprocessing such as drying and H_2S removal (Electrochaea 2014).

CSTR

As previously mentioned, CSTR stands for continuous stirred tank reactor. Commonly used in digesters, it is also used in methanation reactors both ex-situ and in-situ. CSTR uses a mechanical stirrer which effectively increases the k_La of the reactor. However, for the CSTR to be effective, the mechanical stirrer needs a considerable amount of energy. Other problems are the formation of foam in the process liquid which inhibits the process (Rusmanis et al. 2019*)*.

Large-scale methanation is present in the CSTR design. The highest performing CSTR noted in the summary has a MER-value of 800 at 99% CH_4 concentration (Electrochaea 2014). The CSTR used is a pilot-scale methanation plant, which is a part of the “P2G - biocat project” led by Electrochaea. This project is specialized in CSTR methanation. The plant uses a 1 MW electrolyser to support its H_2 injection. Biogas from a wastewater treatment plant is used for CO_2 source. The operating pressure of the reactor is 4-9 bar and it has a temperature of 60-65 °C (Electrochaea 2014). Other authors reported CSTR systems reaching MER values of 285-689 (Rachbauer et al. 2016).

TBR

Trickle bed reactor (TBR) increases its k_La value by maximizing the contact area between gas and liquid. TBR uses a fixed bed, in which the microorganisms are immobilized. A liquid medium containing nutrients is trickled on top of the bed, creating a homogenous distribution of liquid over the bed. H_2 and biogas (or CO_2) are injected at either the bottom or top of the reactor flowing upwards or downwards respectively. The TBR commonly uses a reservoir at the bottom of the reactor where the liquid can be collected and recirculated (Rusmanis et al. 2019). While CSTR requires higher energy input, the TBR is a rather simple reactor with fewer moving parts. Pressure in a TBR can vary, producing high performances in both atmospheric and higher pressure (Strübing et al. 2017; Ullrich & Lemmer 2018). Output gas from a TBR needs postprocessing of drying and H_2S removal (Burkhardt et al. 2019).

The highest performing TBR presented in Rusmanis et al. 2019 has an MER value of 1.5 and a concentration of 98%. The reactor was created at an experimental level and had a packed volume of 5.78 L. The process was done at mesophilic conditions

(37 °C) (Rachbauer et al. 2016). Other reports demonstrated higher-performing reactors, up to MER of 15.4 with CH_4 concentrations above 95% (Strübing et al. 2017). A collection of reported TBR performances is listed in table 1. An important correlation can be noted between MER, HLR and operating conditions. By increasing the HLR of the process so does the performance in MER increase. Two reports regarding mesophilic temperatures with atmospheric pressure show a maximum of 6 HLR. Increasing the HLR would yield a lower concentration of CH_4 (Rachbauer et al. 2016; Burkhardt et al. 2015). However, the HLR can be increased by increased pressure or using thermophilic conditions. (Burkhardt et al. 2019; Ullrich & Lemmer 2018; Strübing et al. 2017).

Table 1: Reported MER, HLR and conditions of trickle bed reactors from literature.

Source	**MER**	**HLR**	**Conditions**
Rachbauer et al. 2016	1.5	6.5	Mesophilic, atmospheric
Burkhardt et al. 2015	1.49	6.0	Mesophilic, atmospheric
Burkhardt et al. 2019	3.11	13.1	Mesophilic, Pressurized
Ullrich et al. 2018	4.28	10.7	Mesophilic, Pressurized
Strübing et al. 2017	15.4*	62	Thermophilic, atmospheric
Strübing et al. 2018	13.1*	52.5	Thermophilic, atmospheric

**Based on reactor bed volume.*

Bubble column

Bubble column reactors or diffusion-based reactors use a fully liquid-based medium where the microorganisms reside. While CSTR uses a mixer to increase the k_La of the reactor, diffusion-based reactors introduce its gas in small microbubbles. A diffusion-based reactor that reaches concentration levels up to 96% at an MER value of 3.7 was reported (Kougias et al. 2017). The system uses in-situ technology at a reactor volume of 2440 m^3. However, no other diffusion-based reactors, ex-situ nor in-situ reaches sufficient CH_4 concentrations as stated by Rusmanis et al 2019.

Other experimental reactors

The “minimal liquid reactor”-type has only been done in experiment levels but shows high performance. The experiment set up reported in Savvas et al. 2017, used a 7-meter-long, 13 mm diameter tube which is hung horizontally in 6 loops. The microorganisms are immobilized at the walls of the tube. Gases and liquid were introduced and flowed through the tube. The k_La was increased by circulating the liquid in intervals giving alternating stages of gas-liquid transfer. By using a thin tube, the active surface area could be maximized. The experiment showed results of an MER value of 40 ($V_{CH4}/V_{reactor}$ * day) and a concentration of 98% CH_4. Except for CSTR, this is one of the highest performing reactors recorded but has only been done at a very low scale with a total reactor volume of 0.75 L (Savvas et al. 2017).

2.3. H_2 production – Electrolysis

Electrolysis of water is the process of using electricity to split water molecules into H_2 gas and oxygen. The H_2 can be considered renewable if the electricity used is from renewable sources. The electrolysis process takes place in an electrolyser cell. The cell contains electrolytes, a membrane, an anode, and a cathode. All components differ for every type of electrolyser but serve the same functions. When an electrical current is introduced, electrons are led through the cathode. At the cathode, water molecules are split into ions and H_2. The ions created from the split are carried over the electrolyte through the membrane to the anode where oxygen is formed. The electrons then transfer into the anode, closing the circuit. The process results in the creation of H_2 gas at the cathode and oxygen at the anode from water. The process is described in formula 2 (Benjaminsson et al. 2013).

$$2\,H_2O \rightarrow O_2 + 2\,H_2 \qquad \text{(Formula 3)}$$

Electrolysers usually operates at higher temperatures because of the three-phase boundary. The three-phase boundary is the meeting point of water, gas, and electrodes where high temperatures lead to higher number of reactions for water molecules. This results in a higher efficiency from electricity to the produced H_2 of the electrolyser. Except for the temperature the efficiency is dependent on the cell’s total electrical resistance from the components of the cell (Benjaminsson et al. 2013).

The electrical efficiency of an electrolyser cell is measured by the energy content of the H_2 produced to the amount of electrical input. The efficiency can be either measured in HHV (higher heating value) or LHV (lower heating value) which can create a difference of 18.2%. For consistency, this study will refer electrolyser efficiency to HHV.

2.3.1. Electrolyser types

Currently, there are four electrolyser technologies that are close to being used or are used commercially: Alkaline (AEL), Polymer electrolyte membrane or Proton exchange membrane (PEM), Solid oxide electrolyser (SOEL), and Anion exchange membrane electrolyser (AEM). Each technology brings advantages and disadvantages and fits different kinds of systems.

Alkaline electrolyser

Alkaline electrolyser or AEL is the oldest and most developed electrolyser type. At the cathode side of the cell, water reacts with the electrons provided from an electrical current and hydroxyl ion (OH^-) and H_2 gas is formed. The ions are transported in the electrolyte, through a permeable membrane. At the anode side OH^- ions lose electrons at the anode where it is converted to oxygen and water (Guo et al. 2019). Due to waters low conductivity of OH^- ions, bases are added. With a higher concentration of bases, the alkaline cell gains electrical efficiency but suffers a loss of life span due to the impact on the cathode and anode (Benjaminsson et al. 2013). Commercial alkaline electrolysers usually use 30%-wt potassium hydroxide solution (KOH) or 25% wt sodium hydroxide solution (NaOH) as electrolytes. Temperatures range typically between 60-100 °C and operating pressure is between atmospheric to 30 bar (Lindorfer et al. 2019). The alkaline electrolyte has a simple design but lacks structural stability. AEL has no expensive materials and therefore has a low production cost (Guo et al. 2019). Electrical efficiency for an AEL is around 70-80%$_{\mathrm{HHV}}$ (Kumar & Himabindu 2019). The expected lifetime of a PEM electrolyser is between 60 000 – 90 000 hours (Schmidt et al. 2017).

Polymer electrolyte membrane electrolyser

The polymer electrolyte membrane electrolyser or PEM has a solid membrane electrolyte. The electrolyte is usually created out of solid poly sulfonated membranes (Shiva Kumar & Himabindu 2019). The anode and cathode are directly connected to the solid electrolyte. The electrolyte works both as a separator membrane and an electrolyte. Deionized water is supplied at the anode, where the water is converted into oxygen gas and releases protons and electrons. The anode process is described in formula 3 (Benjaminsson et al. 2013).

$$2\,H_2O \rightarrow O_2 + 4\,H^+ +\ 4\,e^- \qquad \text{(Formula 3)}$$

The protons together with electrons travel to the cathode through the electrolyte resulting in the separation of oxygen and protons. At the cathode side, protons and electrons react to create H_2 gas, as described in formula 4 (Benjaminsson et al. 2013).

$$4\,H^+ +\ 4\,e^- \rightarrow 2\,H_2 \qquad \text{(Formula 4)}$$

The PEM electrolyser has an operating temperature of 50-80 °C and can have operating pressure up to 85 bar (Lindorfer et al. 2019). It has a higher current density than its alkaline counterpart and has a high conductivity in its electrolytes, which leads to higher efficiency. PEM electrolysers are usually more expensive due to their anode and cathode (catalysts) are made of noble metals due to the acidic nature of the nafion membrane (Guo et al. 2019). Electrical efficiency for a PEM is 80-90%$_{HHV}$ (Kumar & Himabindu 2019). The expected lifetime of a PEM electrolyser is between 20 000 – 60 000 hours (Schmidt et al. 2017).

Solid oxide electrolyser

Solid oxide electrolyser or SOEC has solid ion-conducting ceramics as electrolyte. The electrolyser has operating temperatures of 650-1000 °C and operates up to 25 bars. SOEC can reach up to 92%$_{HHV}$ efficiency which is the highest performing electrolyser. However, it is less developed than both AEL and PEM and is not yet used commercially. Stack lifetime of SOEC is up to 10 000 hours (Schmidt et al. 2017).

Anion exchange membrane

Anion exchange membrane electrolyser or AEM is one of the newest types of electrolysers. As the name suggests, the electrolyser uses an anion exchange membrane as a separator. Water is reduced at the anode side where OH^- is formed similarly to an alkaline electrolyser. The anode and cathode are in direct contact with the separator and OH^- is diffused into the membrane. At the anode, H_2 is produced and OH^- transports to the cathode where oxygen is formed. AEM has a lower operating temperature at 50-70 °C and operates up to 3 bar. Like PEM, the membrane of the AEM is solid making it mechanically stable. While PEM uses nafion membrane, creating the need for noble metals as catalysts AEM does not have this requirement.

AEM electrolysers have the advantage of both stability and cheaper production costs (Vincent 2018). AEM electrolysers are not a mature technology, and the technology is only available for small scale uses. The effective lifetime of the AEM electrolysers is also uncertain (Nohrstedt & Kristensson n.d.).

2.3.2. Electrolyser stacks and electrolyser systems

The type of electrolyser previously mentioned is the different cells or stacks. The stack is the core part of an electrolyser, but other systems need to be in place to have an effective H_2 production. Electricity supply, drying of gas, cell cooling, water circulation and gas purifying all need to be in place and be specified to the electrolyser stack. Suppliers of electrolysers can offer both singular stacks but also a whole electrolyser system. The benefits of stacks are that the user can customize the system at a high level. Electrolyser systems do not often bring this sort of freedom but bring a simpler solution. Small-scale or even medium-scale electrolysers can be positioned virtually anywhere, provided the connection of water and electricity supply is available. Larger electrolysers can come as “container solution” meaning all equipment and electrolyser are installed in a container that can be positioned and installed at a site.

2.4. Costs of BHM

The production costs of H_2 and CH_4 is a well-documented area. The benefits of H_2 and CH_4 for substituting fossil fuels are prevalent and there is a large interest in power to gas (P2G). Though studies of small-scale production of CH_4 from methanation are limited. Production costs for systems with <500 kWe are not well investigated and more commonly costs of plants up MW scale are documented. Specific investment costs can be estimated following the “six-tenth-factor rule” which is presented in equation 1 (Zauner et al. 2019).

$$C_b = C_a * \frac{S_b}{S_a}^f \tag{1}$$

Where C_b is the cost of the questioned system, C_a is the cost of the same type of system but at different scales and S_b and S_a are their respective scales. f is the scale factor of the system, which differs for different technologies and systems.

Both electrolysers and methanation systems are still new to the market and when investigating the costs of production, it is important to understand the learning curve concept to see potential in future investments. Manufacturing processes are optimized in a competitive environment. The concept of the learning curve is that experience of manufacturing a specific technology will increase over time and in turn creating lower costs of the technology (Böhm et al. 2018).

2.4.1. Production cost of H_2

The production costs of H_2 depend on electrolyser type and scale. A study of specific CAPEX per capacity of electrolyser (€/kWe) was performed, in which PEM, AEL, and SOEC were analysed. A standardized specific CAPEX for a 5 MW electrolyser plant was calculated from different sources, and the results of this study are shown in table 2 (Zauner et al. 2019). Similar results can be seen in other studies (Christensen 2020). Previous literature indicates that PEM has higher material costs than AEL (Schmidt et al. 2017). However, Christensen 2020 mentions other systems such as compressors are not required for PEM which can reduce the costs of the H_2 production for that specific type. Specific investment costs from the two different studies are shown in table 2.

Table 2: Estimated investment costs for different types of electrolysers from literature. Top row costs are based on a scale of 5 MW_e (Conversion: 1 \$ = 0.82 €)

	AEL	**PEM**	**SOEC**	**Source**
Production cost (€/kWe)	1060	970	1990	Zauner et al. 2019.
Production cost (\$/kWe)	988 (814 €/kW)	1182 (973 €/kW)	1346 (1108 €/kW)	Christensen 2020.

Electrolyser OPEX costs (such as water and operation of necessary equipment) are estimated at 1-3% of the CAPEX for the electrolyser. The electrolyser lifetime also has a great impact on the production costs of H_2. Electrolyser replacements can be estimated to 25 - 50% of original electrolyser CAPEX (Christensen 2020; McDonagh et al 2018). Future reduction of H_2 production costs due to the concept the of learning curve is estimated at 2 %/year (Christensen 2020) or 28-35% in the year 2030 (Zauner et al. 2019).

Although the studies do not cover many small-scale electrolysers, an average scale factor of 0.75 was acknowledged (Zauner et al. 2019). Calculated specific CAPEX using for AEL and PEM at lower scales using equation 1 and values presented in table 2 can be seen in figure 3.

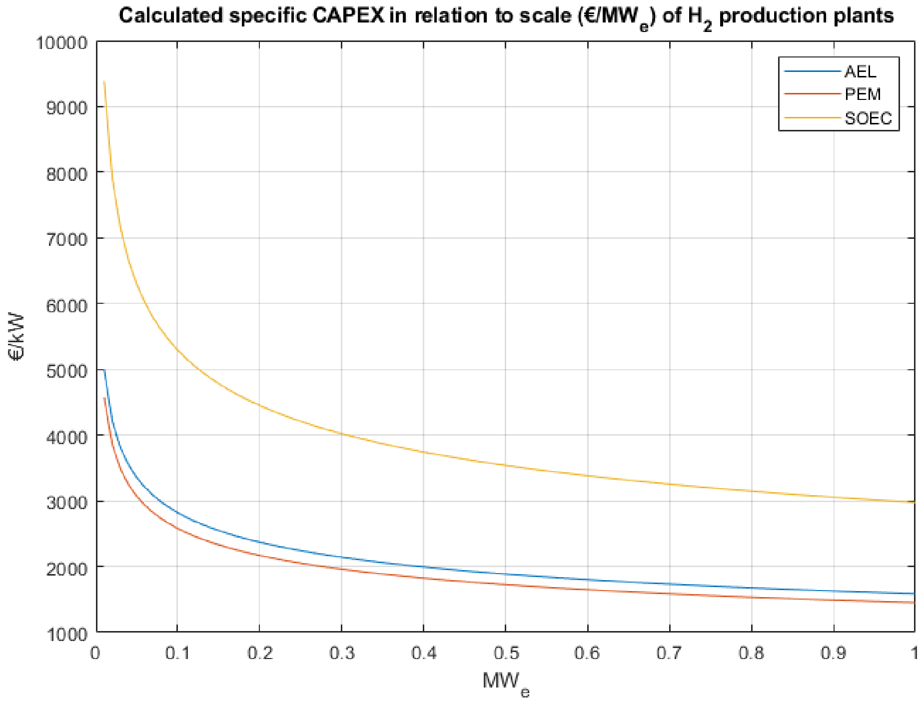

Figure 3:Calculated specific CAPEX for AEL, PEM and SOEC based on calculation data from Zauner et al. 2019.

During the present study real CAPEX of electrolysers were gathered for scales below 500 kW. These costs are presented in Appendix 1.

2.4.2. Production cost of CH_4

Investment costs for a biological methanation plant differ on the scale of the plant. Investment for a BHM plant includes a reactor, electrolyser, engineering, machinery, and commissioning. The relation between these factors, changes depending on the scale of the plant. In figure 4 and 5, it can be observed that a higher percentage of total CAPEX can be attributed to the electrolyser, as the plant increases in scale (Electrochaea 2014).

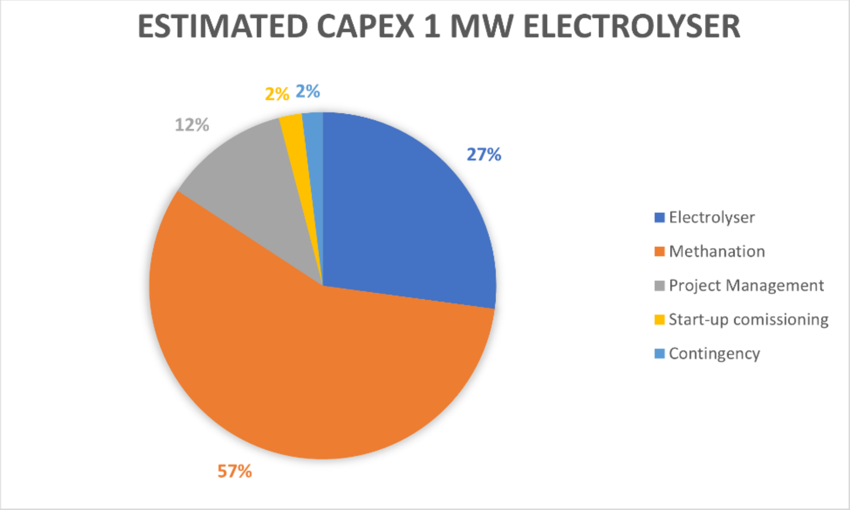

Figure 4: The estimated investment cost factors of a biological methanation plant using a 1 MW electrolyser. Methanation (orange) are the machinery and reactor for the methanation. Source: Electrochaea 2014.

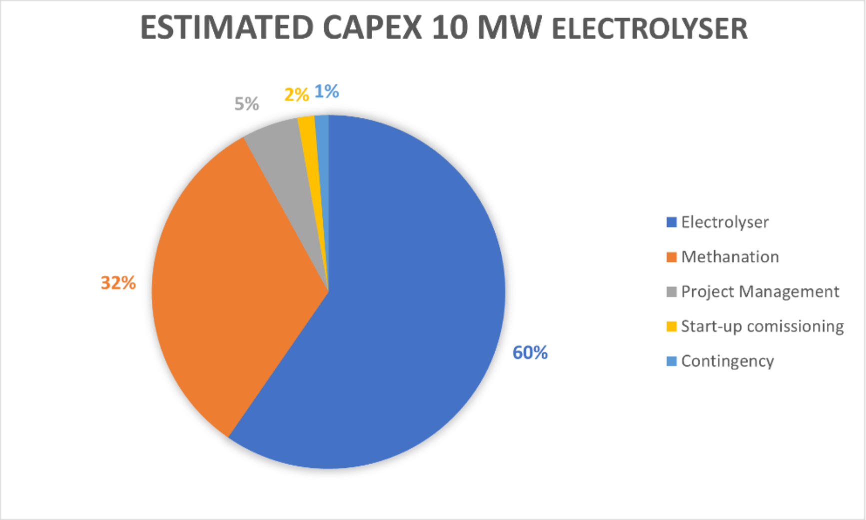

Figure 5: The estimated investment cost factors of a biological methanation plant using a 10 MW electrolyser. Methanation (orange) are the machinery and reactor for the methanation (Electrochaea 2014).

Reports performed by Zauner et al. 2019 show estimated specific investment costs between 90-1800 €/kW$_{SNG}$ for different BHM plants. It is based on different 8 sources and the scale of the plants ranges from 200 kW to 50 MW. The smallest scale investigated at 200 kW production of CH_4 have a specific investment cost of 320 €/kW. The same study also estimated 1 MW production to 120 €/kW while other references report up to 1440 or 1800 €/kW for the same rated production of CH_4. According to the information in 2019, the standardized price of a 5 MW plant has the specific investment cost of 600 €/kW with a scale factor of 0.52 which is visualized in figure 6 by calculations done with equation 1.

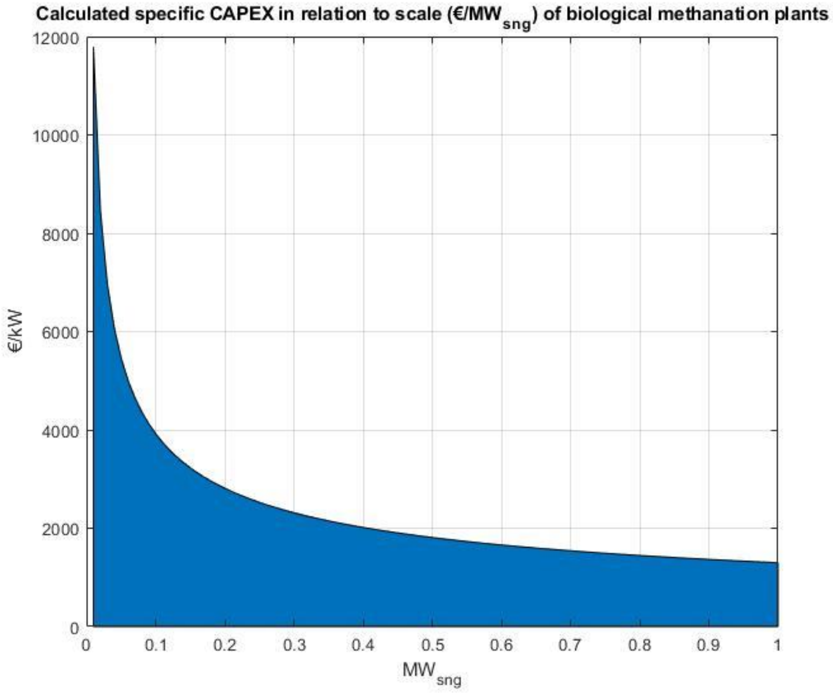

Figure 6:Calculated specific investment costs based on 600 €/kW for a 5 MW plant with a scale factor 0.52. The figure shows the calculated costs between 0 and 1 MW$_{sng}$ scale.

With the specific investment cost of 600 €/kW for 2019, a potential reduction of 35% can be expected by the year 2030 (Zauner et al. 2019). Usually, excess heat is generated at both electrolyser and reactor. This creates a potential heat recovery that can substitute heating costs which can provide a BHM system with additional revenue.

2.4.3. Climate Leap initiative

Climate Leap initiative (Klimatklivet) is a subsidy program in Sweden to lower greenhouse gases. The program is aimed at solutions both locally and regionally. The Climate Leap initiative is set to allocate 2.3 billion SEK in 2021 and is led by Naturvårdsverket and Länsstyrelserna. Subsidies can be given to different organizations such as companies or municipals which provide solutions to reduce greenhouse gas emissions in energy, transport, infrastructure, or industry sectors. The maximum subsidy in the Climate Step Initiative is 50% of the total investment costs of the selected solution (Natruvårdsverket 2021). Biogas production and methanation has a large opportunity to gain this subsidy and can help develop methanation as a solution to lower greenhouse emissions.

3. Method

To simulate energy balance, mass balance and production costs of a concept BHM plant at four scales a system description with boundaries and chosen technology was designed. Each case of scale is defined by the installed capacity of electrolyser. The cases are 4.8 kWe, 20 kWe, 100 kWe and 550 kWe.

Choice of technology for each case was based on literature and economical study performed at the start of the project. A MATLAB model was designed to handle all calculations necessary and is based on the system description. The system boundaries, motivation to technologies, system description and model breakdown is presented in this chapter.

3.1. System boundaries

The concept system is assumed to be used in as part of a biogas upgrading process. Therefore, the system will only contain the methanation process, H_2 generation, nutrient supply, and H_2S removal process. The following assumptions are made for the system:

- Biogas is assumed to be supplied from a digester with 60% CH_4 concentration at 10 °C.
- A part flow of digestate is supplied to the system at 15 °C from the digester.
- Produced O_2 is not considered in the scope of the system. O_2 assumed to be injected into the digester to lower H_2S concentration of output gas after methanation is assumed to be 50 ppm.
- The ambient temperature is assumed to be 20 °C for reactors located inside.
- Excess heat that can be recovered (for electrolysers using liquid cooling) is used for preheating substrate for the digester. It is assumed that this lowers the temperature of the cooling water to 15 °C.

- The lifetime of the system is estimated to 20 years and 8500 hours of yearly operation. Electrolyser lifetime is assumed to be 7 years which results in the need of two electrolyser replacements.
- No degradation of electrolyser is assumed. However, the replacement of electrolysers is accounted for.
- OPEX for nutrient supply was not considered due to missing data.

3.1.1. Digester size

Because it is hard to understand or visualize the scale of the system by electrolyser capacity, it is important to put the cases of scale into to perspective. In table 3, the digesters calculated yearly production and biogas flow are shown. The table shows the size of a biogas plant which the system can serve with upgrading. The calculations are based on equation (2) (which is presented in 3.4.2), the assumed concentration of CO_2 in the supplied biogas and the H_2 production for each case (table 4).

Table 3: Calculated biogas flow and production from digester based on electrolyser capacity.

	4.8 kWe	**20 kWe**	**100 kWe**	**550 kWe**
Yearly injected biogas (MWh/y)	34	122	738	3760
Biogas flow (Nm^3/h)	0.7	2.4	14.5	73.9
H_2 production (Nm^3/h)	1	3.64	22	112

For perspective, the yearly biogas production ranges between 300 to 8000 MWh/y based on 32 farm-based biogas (Eliasson 2015). It can be noted that cases 4.8 and 20 kWe has a lower production than the smallest scale of the evaluated biogas plants. It is assumed that these scales upgrade a part flow of a biogas plant, while 100 and 550 kWe cases can upgrade a full flow of biogas.

3.2. Motivation for choice of technologies

BHM plants can use different types of reactors, electrolysers, and post-processing to achieve the same goal. The choices of these technologies are described with motivations as to why and what alternatives could be used. Technology choice may also differ for the cases investigated due to scale.

3.2.1. Electrolyser

The choice of electrolyser for the system is AEM, PEM and AEL, depending on the case. Different suppliers were contacted for pricing and electrolyser specifics. An overview of the electrolysers used is shown in table 4.

Table 4: Electrolyser specifics for each chosen electrolyser. Specifics are based on spec sheets for each electrolyser.

	4.8 kWe	**20 kWe**	**100 kWe**	**550 kWe**
Supplier	1	2	2	3
Electrolyser type	AEM	AEL	AEL	PEM
H_2 production	1 Nm^3/h	3.64 Nm^3/h	22 Nm^3/h	112 Nm^3/h
Efficiency $\%_{HHV}$	74%	65%	78%	72%
Water consumption	0.8 L/h	2.5 L/h	16 L/h	170 L/h
Cooling type	Air	Liquid	Liquid	Liquid

Case 4.8 kWe

For this scale, the system uses two AEM electrolysers based on supplier 1. Supplier 1 uses a highly adaptive modular design, where several electrolysers can easily be integrated. These electrolysers were chosen before both AEL and PEM due to their high efficiency at such a small-scale ($74\%_{HHV}$) and a low investment cost. The estimated investment cost for this electrolyser was gathered from supplier 1. Another benefit AEM provides is that the system does not need purified water or any KOH solution for operation, avoiding extra investment and operational costs. The disadvantage of this electrolyser choice is the AEM does not use liquid cooling, making it difficult to recycle waste heat which AEL and PEM electrolyser can do. However, at this scale, small amounts of heat are released.

Case 20 kWe and 100 kWe

For both cases at 20 kWe and 100 kWe, AEL is used and are based on alkaline electrolysers from supplier 2. AEL requires both purified water and KOH solution to operate, which must be added to the system. AEL is used in these cases because of high efficiencies ($70\text{-}79\%_{HHV}$). Other PEM electrolysers investigated at this scale had lower system efficiency than AEL. Still, PEM may be a good option for this scale. AEM was first suggested to be used at 20 kWe due to the highly modular design and stacking multiple 2.4 kW AEM electrolysers. However, the AEL is liquid-cooled and enables waste heat recovery. It is important to note that the exact CAPEX for these two electrolysers is estimated due to a lack of data.

Case 550 kWe

Two 225 kW PEM electrolysers based on supplier 3 were chosen. The electrolysers use a container solution. Both AEL and PEM were considered at this scale. Based on reports detailed in 2.5.1 AEL and PEM at this scale have similar production costs. PEM however has a compact design, shorter start-up and a high system efficiency of $74\%_{HHV}$. The estimated cost for this electrolyser was also gathered from supplier 3, giving the estimated investment cost of this electrolyser larger accuracy. The electrolysers include purifying, only water and electricity need to be added which removes the need for KOH solution if AEL was used.

3.2.2. BHM reactor

The chosen reactor for all cases is a trickle bed reactor. Due to the high CH_4 concentration of output gas and considerable high MER values, CSTR was considered for the system and could be a good option. However, due to the complex nature of high pressure and energy usage from the CSTR mixer, TBR was chosen over CSTR. Other reactors such as minimal liquid and diffusion-based reactors were also considered but ultimately discarded due to the limited amount of literature data on these reactors performing at satisfying levels. Numerous pieces of literature on TBR report high performance with a concentration above 95% CH_4 (Table 1) and the concepts for the reactor follow a similar setup of the reactor reported in Strübing et al. 2017. The reactor requires a buffer to keep pH levels from declining. The decline of pH is due to high HLR, and irregular flows may disrupt production (Strübing et al. 2017).

3.2.3. Nutrient supply

The necessary nutrients that need to be supplied for the reactor are assumed to be taken from the liquid phase of the digestate exiting the digester. From a dewatering process, the reject (water phase) is estimated to contain enough nutrients to supply the reactor. The method of extracting the liquid is dependent on scale. For cases at the scale of 4.8 kWe and 20 kWe, filtration bags are used. For the larger systems at scales of 100 kWe and 550 kWe which have higher nutrient flow, a screw press is used for dewatering the digestate.

3.2.4. H_2S removal

If the output gas is to be considered as fuel it needs to fulfil the requirement of low concentrations of H_2S. For H_2S removal an activated carbon filter module is used for all scales. Activated carbon was chosen both for its costs and for its high efficiency of removing H_2S from product gas.

3.3. Concept system description

The basic description of the system is shared for all four cases. A system overview is illustrated in figure 7. The design of the system is based on various literature.

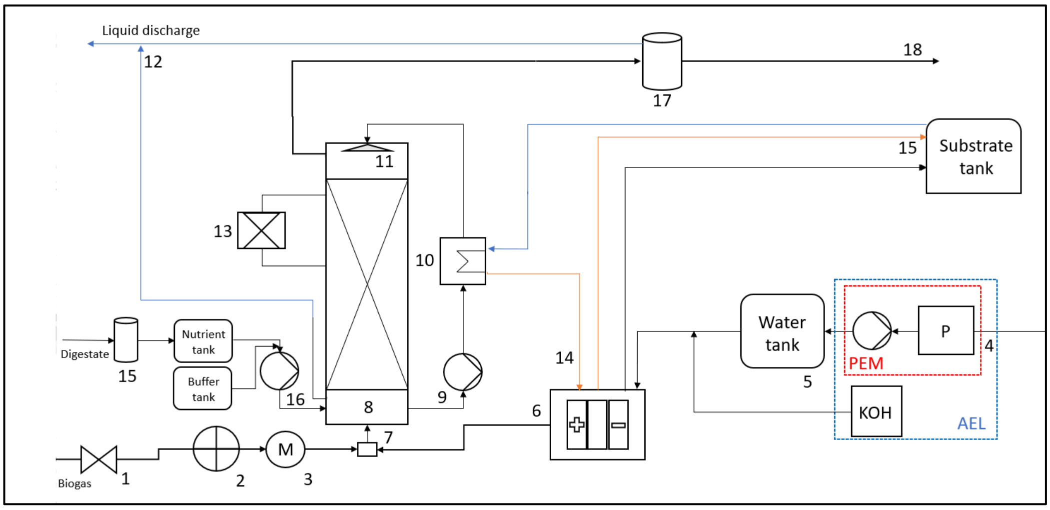

Figure 7: Schematic diagram of the system. The illustration details gas, liquid and cooling flow. The schematic shows a shut-off valve (1), biogas fan (2), flow meter (3), purifier/KOH setup(4), water tank (5), electrolyser (6), injection valve (7), reactor reservoir (8), circulation pipes with pump (9), heat exchanger (10), spraying nozzle (11), liquid discharge line (12), measurement equipment (13), cooling line to digester (14, 15), filtration system (15), nutrient and buffer injection (16), H2S removal with activated carbon (17) and output gas (18).

Supply of biogas

As previously mentioned, it is assumed that the system receives a supply of biogas from a nearby digester. The biogas is cooled to a ground temperature causing vapour in the gas to condense and excess water is discharged before intake. The main shut-off valve is installed, where the flow of biogas can be completely stopped if necessary (1). After the valve, a gas fan is used to lead the gas to the reactor (2). A flowmeter is positioned after the fan to measure the intake of biogas (3).

H_2 generation

Electrolyser setup is designed similarly to Guo et al. 2019 and is used for AEL electrolysers. For cases using AEL, tap water is fed into a purifier (4). The water is then supplied into a pressurized water tank (5) for all cases. Subsequently, the water is injected into the electrolyser/s. H_2 generated at the electrolyser (6) is injected into the reactor together with biogas (7). The oxygen is assumed to be injected into the digester.

Reactor

The reactor size depends on the case of the system, but operating procedures follow the same principle. The TBR is operating at a temperature of 55 °C and close to atmospheric pressure. Because of thermophilic conditions, it is assumed that the reactor has an HLR of 62 $V_{h2}/(V_{reactorbed} * day)$. The TBR has a nutrient reservoir at the bottom of the reactor (8). Nutrients are injected into the reservoir. A circulation pipeline is connected to the reservoir (9) leading to the top of the reactor. The circulation pipe is located at the outside of the reactor where it is connected to a heat exchanger (10) which heats the circulation liquid to a higher temperature. The liquid medium is then sprayed by a nozzle at the headspace of the reactor (11). The circulation liquid is used for heating the reactor and no liquid heating loop is installed into the reactor shell. The liquid medium flows down through the bed, delivering nutrients to the methanogens. The liquid is then returned to the reservoir and is circulated again. The reservoir is designed to discharge the overflowing liquid (12). The temperature, pH and pressure of the reactor are measured (13).

Heat recovery

The reactor and electrolyser require cooling, and a liquid cooling line is used. The cooling liquid used is water. The cooling process begins at the reactor, where the circulation pipes of the reactor are connected to a liquid heat exchanger (10). The cooling liquid cools the circulation water. The cooling line connects to the electrolyser (14), when liquid cooling is applied. If the electrolyser is small, it uses air cooling and will not be cooled by liquid. After passing the electrolyser, the heated cooling liquid is transported to a substrate holding tank to preheat substrate (15). The cooling liquid is assumed to return to the heat exchanger at 15 °C.

Nutrient supply

A fraction of used digestate is drawn from the digester outlet. The digestate is drained of water through a water drainage process (15). For 4.8 and 20 kWe systems the flow of nutrients is so low that simple filters changed every 4 days can support the system. For 100 and 550 kWe systems, the flow is larger, and a screw press is used to extract the nutrient medium. In either method, the nutrient medium is stored in a tank. Periodically nutrients are supplied together with a buffer solution (K_2HPO_4) to the reactor reservoir (16).

H_2S removal

The output gas from the TBR is then transported to H_2S removal. First, the gas will be cooled to 8 °C and excess condense will be removed before the H_2S removal. The gas is then injected into an activated carbon filter (17). The module will remove a large part of the H_2S. The gas is then led out of the system (18). The output gas of the system is a high CH_4 concentrated biogas with removed H_2S but it is still water-saturated and a drying process is needed for full vehicle grade biogas.

3.4. Model

3.4.1. Model inputs

Inputs for the model are shown in table 5 listing electrolyser specifications and reactor operation conditions. Electrolyser inputs are based on real commercial electrolysers. Inputs for these are nominal power usage, water consumption, specific production, and cooling requirements. All electrolysers are operated at 60 °C. Reactor operation inputs are the same for all cases and are operating temperature, conversion rate, H_2/CO_2 ratio, HLR and whether the reactor is placed outside or inside depending on scale. If the reactor is placed outside, it will have a thicker insulation.

Operational parameters of the system such as full load hours (FLH) for one year and the total lifetime of the system are set to 8500 h/y and 20 years, respectively. The system is designed to operate at full time with the digester. Electrolyser lifetime depends on type. However, all electrolysers have an estimated lifetime of 60 000 hours. At operating conditions, the electrolysers are estimated to operate fully for 7 years, resulting in electrolyser replacements of 2 for the lifetime of the plant. Biogas that will be upgraded is set to have a concentration of 40%-vol CO_2 and 50 ppm of H_2S. The activated carbon filter needs to be refilled, preferably as few times as possible. It is estimated to be refilled 2 times per year.

Table 5: Model inputs for the mass balance, energy balance and production of the simulation.

Input	4.8 kWe	20 kWe	100 kWe	550 kWe
Electrolyser specifics*				
Supplier*	1	2	2	3
Electrolyser type	AEM	AEL	AEL	PEM
H_2 production	1.0 Nm^3/h	3.6 Nm^3/h	22.0 Nm^3/h	112 Nm^3/h
Water consumption	0.8 L/h	2.5 L/h	16 L/h	170 L/h
Cooling method	Air	Liquid	Liquid	Liquid
No. of replacements**	2	2	2	2
Reactor operation***				
Operating temperature	55 °C			
HLR	62 (V_{CH4}/V_{rb}*day).			
H_2/CO_2 - ratio	3.76			

Conversion rate	98%			
Placement	Inside	Inside	Outside	Outside
Operation				
FLH per year	8500 h			
Lifetime of system	20 years			
Biogas properties				
Concentration of H_2S	50 ppm			
Concentration of CO_2	40%-vol			
Activated carbon				
Refills/year	2			

**The inputs of the electrolyser are based on spec sheets for each electrolyser.*

***Estimated lifetime for each electrolyser is 60 000 hours, meaning 2 replacements are needed for 20 years of service.*

****Reactor operation inputs are based on the set up from Strübing et al. 2017.*

3.4.2. Energy and mass balance calculations

Constants for gas, liquid and material properties are presented in Appendix 3. The estimated production of the system is calculated through several steps. First, the estimated amount of CO_2 injected into the reactor (n_{CO2}) is calculated by:

$$n_{CO2} = \frac{n_{h2}}{r_{H2/CO2}} \ (mol/h) \tag{2}$$

Where n_{h2} is the amount of injected H_2 (mol/h) and $r_{H2/CO2}$ is the ratio between injected H_2 and CO_2 (3.76). The amount of CH_4 generated due to methanation is then calculated by:

$$n_{CH4} = n_{CO2} * 1 - r_{bound} * r_{conversion} \tag{3}$$

Where the n_{CH4} is the amount of generated CH_4 in mol/h, r_{bound} is the amount of CO_2 that is absorbed by microorganisms (6%) and $r_{conversion}$ is the conversion rate of the methanation process (98%). With equations 2 and 3 and gas attributes such as density and LHV, the production of the system can be calculated.
The volume of the reactor is calculated by:

$$V_{rb} = \frac{P_{H2} * 24}{HLR} \tag{4}$$

HLR is the specific H_2 loading rate (62 $Nm^3{}_{H2}$ / $m^3{}_{Rb}$, day), P_{H2} is the H_2 production (Nm^3/h) and V_{rb} is the volume of the reactor bed (m^3). Headspace and reservoir are assumed to be 10% each of the bed volume. The sum of headspace, reservoir and bed is the assumed total volume of the reactor.

For the mass balance, all key figures are based on the small-scale TBR in Strübing 2017. Key figures of the mass balance are:

- Specific nutrient flow (13 L/(h*m^3) - The amount of nutrients needed per volume of reactor bed.
- Specific buffer flow (0.07 mL/(h*m^3) – The amount of buffer needed per volume of reactor bed.
- Circulation flow (172 L/(h*m^3) – The flow of the liquid circulation in the reactor.

By using these key figures and the calculated volume of the reactor, the nutrient, buffer, and circulation flows can be calculated.

The mass balance of water in the reactor is then calculated by:

$$\dot{V}_{water} = \dot{V}_{buffer} + \dot{V}_{nuntrient} + \dot{V}_{gas\,in} + \dot{V}_{meth} - \dot{V}_{gas\,out} \tag{5}$$

Where $\dot{V}_{water}$ is the flow of liquid out of the reactor (L/h), $\dot{V}_{buffer}$ is the added buffer (L/h), $\dot{V}_{nuntrient}$ is the added flow of nutrients (L/h) and $\dot{V}_{meth}$ is the amount of water that is generated inside the reactor following formula 1. $\dot{V}_{gas\,in}$and $\dot{V}_{gas\,out}$is the amount of water added or removed in the reactor by gas (L/h) and is calculated by gas mass fraction of 1.8% and 8% respectively.

For calculating an estimation of heat transmission losses, the reactor is simplified to a cylinder with a height to radius ratio of 5. The reactors heat transmission heat losses are then calculated by:

$$Q_T = A\frac{T_1 - T_2}{R} \tag{6}$$

Where Q_T is transmission losses (W), A is the surface area (m^2), T_2 is reactor operating temperature (55 °C), T_1 is the ambient temperature (°C) and R is the heat resistance of the reactors shell. The resistance is calculated by:

$$R = \frac{1}{h_1} + \frac{1}{h_2} + \sum \frac{L_i}{k_i} \tag{7}$$

Where h_1 and $h_2$is the convection of the shell's surfaces (50 W/m^2°K and 500 W/m^2°K respectively), L_i is the thickness of each layer of the shell (m) and k_i is the heat transfer coefficient of the layer (W/m^2°K).

Heat losses from injected liquid and gases are calculated by:

$$Q_L = \dot{m}\, c_p\, \Delta T \tag{8}$$

Where Q_L is the amount of energy the gas/liquid is needed to heat up to reactor temperature, $\dot{m}$ is the mass flow of liquid/gas (kg/s), $c_p$is the specific heat capacity of the gas/liquid (W/°K) and ΔT is the temperature difference between ambient and reactor temperatures (°C). The ambient temperature is either set to 20 for systems inside (4.8 kW and 20 kW) while varying temperature based on 2019 hourly data (SMHI 2021) is used for systems outside (100 kW and 550 kW). H_2 gas injected provides heat to the reactor by the same equation.

Due to the methanation reaction is exothermic the heat generated from the methanation process is calculated by:

$$Q_m = n_{CH4} * E_m \tag{9}$$

Where Q_m is the energy amount generated from methanation (kJ/h), n_{CH4} is the amount of converted CH_4 (mol/h) and E_m is the energy generated per mol from formula 1 (167 kJ/mol). It is assumed that all energy generated is heat.

The amount of heat generated from the electrolyser is calculated by:

$$E_{elheat} = P_{el}\, f_{heat} \tag{10}$$

Where E_{elheat} is the excess heat from the electrolyser, P_{el} is the input electricity of the electrolyser and f_{heat} is the fraction of the input energy that is excess heat and is set to 17% for operating temperatures of 60 °C (Frank et al. 2018).

KOH consumption for AEL electrolysers is calculated by (Kuckshinrichs et al. 2017):

$$m_{KOH} = m_{H2} * 2.75 * 10^{-4} \tag{11}$$

Where m_{KOH} is the amount of KOH (kg/h) and m_{H2} is the amount of H_2 produced (kg/h). Estimated electricity need for pumps are calculated by:

$$P_p = \frac{\dot{m}_V g\, \Delta p}{\eta} \tag{12}$$

Where P_p is the power usage of the pump, $\dot{m}_V$ is the mass flow of liquid (m/s), g is the gravity constant (9.81 m/s^2), Δp is the pressure difference and η is the efficiency of the pump (90%). Finally, electricity usage of the gas fan is assumed to be 7.5 W/Nm^3 biogas.

3.4.3. Economic assessment

The estimated CAPEX of the system is shown in table 6. Costs for the reactor, equipment, installation, ground preparation and commissioning are based on discussions with the current study's project manager at RISE[1]. Electrolyser costs are based on the current study's economic analysis. The electrolyser replacement costs are estimated at 40% of electrolyser CAPEX (McDonagh et al. 2018). Case 4.8 kWe and 550 kWe uses real CAPEX costs gathered from supplier 1 and 3, respectively. Case 20 kWe and 100 kWe have estimated electrolyser costs based on costs for AEL electrolysers shown in table 2. OPEX factors of electricity use, water use, activated carbon, filter capsules, reparations, staff costs, buffer, and KOH solutions are also presented in table 6.

Table 6: Investment and operational costs for each case.

CAPEX/OPEX factor	**4.8 kW$_e$**	**20 kW$_e$**	**100 kW$_e$**	**550 kW$_e$**	**Source**
CAPEX					
Electrolyser (€/kW)	3600	3500*	2751*	2363**	-
Electrolyser replacement cost (%)	40	40	40	40	McDonagh et al. 2018
Reactor (€/m^3)	19200	11000	9500	3500	Estimation
Other equipment and installation (€)	85000	100000	150000	200000	RISE
Ground preparation and commissioning (€)	20000	20000	40000	50000	RISE
OPEX					
Electric price (€/kWh)	0.059				Estimation
Buffer (€/kg)	80				Sigma-Aldrich 2021
KOH (€/kg)	2.5				Kuckshinrichs et al. 2017

---

[1] Johan Andersson, Senior project manager, RISE, personal meeting, 2021-5-25

Activated carbon (€/kg)	172	RISE
Purifier capsules (€/capsule)	105	Vattensystem
Water (€/L)	0.0049	Uppsala vatten 2021
Staff cost (€/h)	30	Estimation
Reparation (% of CAPEX)	2	Estimation

**Estimated costs.*

***PEM electrolyser includes purifier and other equipment.*

Economic results of the simulation are presented as levelized cost of CH_4 production (LCOCH4, €/kWh) and is calculated by:

$$LCOCH4 = \frac{\frac{\sum_{t=1}^{n} CAPEX_t + OPEX_t}{(1+r)^t}}{\frac{\sum_{t=1}^{n} E_{CH4}}{(1+r)^t}} \tag{13}$$

Where E_{CH4} the production of CH_4, t is the current year, n is the lifetime of the plant, $CAPEX_t$ is the investment costs for the year, $OPEX_t$ is the operational costs for the year, and r is the discount rate (5%). 5% of the total CAPEX is also added in OPEX as return of investment and interest on investment loan. Another economical parameter is the investment costs per capacity of the plant (COCH4) in €/kW and is calculated by:

$$COCH4 = \frac{\sum CAPEX_i}{P_{CH4}} \tag{14}$$

Where P_{CH4} is the rate of production for the plant (kW_{CH4}) and $CAPEX_i$ is the investment costs for the plant. This includes all CAPEX presented in table 5.

The model also calculates recovered heat. An additional calculation of LCOCH4 and COCH4 is calculated for each case to investigate the effects of using the recovered heat. Recovered excess heat provided from both reactor and electrolyser is assumed to substitute heating from a wood chip pan. This cost is estimated to 0.04 €/kWh.

Upgrading costs and BHM production costs

Both LCOCH4 and COCH4 depend on the systems production of CH_4. However, the production of CH_4 changes depending on if the CH_4 injected by biogas is accounted for. Therefore, the results will be presented in two ways:

- $LCOCH4_{Total}$ and $COCH4_{Total}$ - The levelized cost of CH_4 and specific investment cost for the total CH_4 in the biogas (including injected CH_4).
- $LCOCH4_{Meth}$ and $COCH4_{Meth}$ - The levelized cost of CH_4 and specific investment cost for the CH_4 produced by the BHM process (excluding injected CH_4).

$LCOCH4_{Total}$ can then be defined as upgrading costs of the biogas while $LCOCH4_{Meth}$ can be defined as the production cost of CH_4 as a methanation system.

Sensitivity analysis

The sensitivity analysis investigates the effects reduction in CAPEX and different operating parameters. A total CAPEX reduction of 35, 50 and 82.5% were investigated for the purpose of investigating effects on the system results of a future cost reductions in 2030, subsidy, or both, respectively. An electrolyser CAPEX reduction of 28 and 45% were performed to investigate the effects of possible CAPEX reduction for the year 2030 for AEL and PEM. Operation hours during the year and electricity price are also varied to investigate effects on the simulation results.

4. Results and discussion

The results of the model simulations are presented and discussed in this chapter. The system performance, energy balance, mass balance, production costs and sensitivity analysis are detailed below.

4.1. System performance

The performance of the system is shown in table 7. The production of CH_4 due to methanation ($P_{methanation}$) and the total production of CH_4 (P_{total}) is shown. The plant produces an output gas with high concentrations of CH_4 (95.7%) with H_2S removal. Only drying of the gas is required for the output gas to reach vehicle fuel grade. The system does also increase the CH_4 yield by 60% for all cases. With an estimated conversion rate of 98% and an estimated 6% of CO_2 bound by biomass the resulting MER value is 15.2 for all cases. This is close to the MER value of 15.4 reported by Strübing (Strübing et al. 2017), which the present model is based on. The system efficiency (kW electricity input/ kW $P_{methanation}$) can be correlated to the efficiency of the electrolyser as shown in table 4 and 7. Cases using higher-performing electrolyser results in higher system efficiency. Therefore, optimizations of H_2 production are of great importance for the efficiency of the system. This correlation can be explained by the high electricity utilization of the electrolyser, as demonstrated in the energy balance calculations.

Table 7: Simulation results of plant performance for each case. The calculated size of the reactor is also presented which increases with scale of electrolyser.

Parameters	**4.8 kWe**	**20 kWe**	**100 kWe**	**550 kWe**
$P_{methanation}$ (kW)	2.4	8.9	53.6	272.9
P_{total} (kW)	6.4	23.2	140.5	715.0
System efficiency (%)*	51	44	54	50
Reactor bed volume (m^3)	0.4	1.4	8.5	43.4
CH_4 (%-vol)	95.7			
MER (V_{CH4}/V_{rb}*day).	15.2			

*System efficiency is measured by: Power of produced CH_4 (kW) devided by total electrical input (kW).

The advantages of this type of system are several. TBR’s have been proven to be able to start production quickly and effectively which is beneficial at irregular biogas production (Strübing et al. 2017). With the use of a TBR, no mixer is required which is less energy intensive. With fewer moving parts in the reactor, it can also be assumed that there will be less downtime due to maintenance which as mentioned in 2.1 is a large economic burden for many small-scale biogas plants (Eliasson 2015). Disadvantages are the lower MER value obtained for TBR compared to CSTR. Continuous monitoring is necessary for the process to run effectively. Fluctuations in biogas, H_2 or buffer supply are apparent factors that can disrupt the production.

4.1.1. Energy balance

An overview of the energy balance of the system is illustrated in figure 8 for case 20 kWe. All cases follow closely the same energy balance percentagewise except case 4.8 kWe, which does not use heat recovery for the electrolyser. Detailed energy and mass balance can be found in Appendix 2. It can be noted that close to all electricity utilization is attributed to the electrolyser and only 0.1 to 1% is used by other equipment. It is important to note that the electricity usage of the plant infrastructure is only attributed to pumps and fans. Lighting, measurement, and control are not considered in this calculation. Similar electricity usage can be seen in a CSTR-based methanation plant, rated at 1 MWe where 97% of the total electricity is to support its electrolyser (Lardon et al. 2018).

Concerning the heat balance both the electrolyser and the reactor generate heat. Due to the exothermic nature of methanation (formula 1), after starting up, the reactor will heat itself. Cooling is then required for both the electrolyser and the reactor. However, some heat losses are prevalent in the reactor. Heating injected gas/nutrient medium and transmission losses of the reactors shell are the major sources of heat loss of the system.

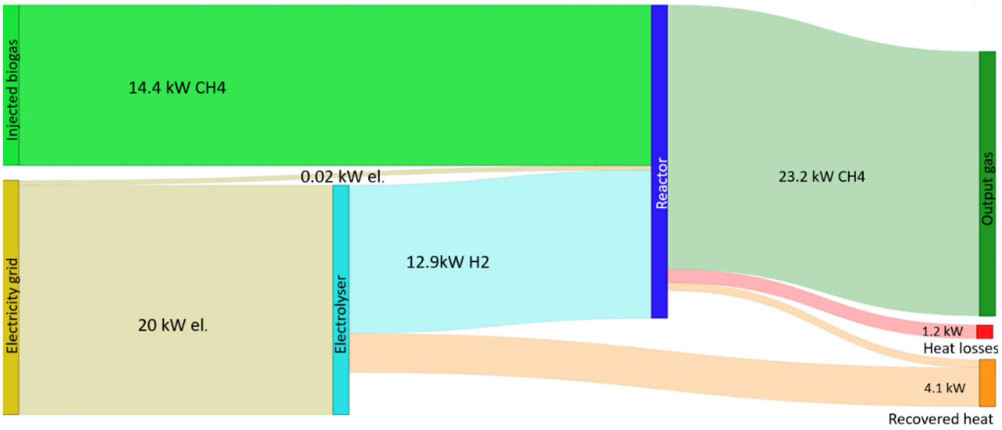

Figure 8: Illustrated representation of the energy balance of the system for case 20 kWe.

4.1.2. Mass flow

The mass flow of the system is illustrated in figure 9. The electrolyser has a water consumption. The injected nutrient and buffer medium increases with reactor size. The liquid medium increases after the reactor due to the water that is generated in the methanation process (formula 1). Both injected H_2 and CH_4 is shown. A small fraction of H_2 remains in the output gas.

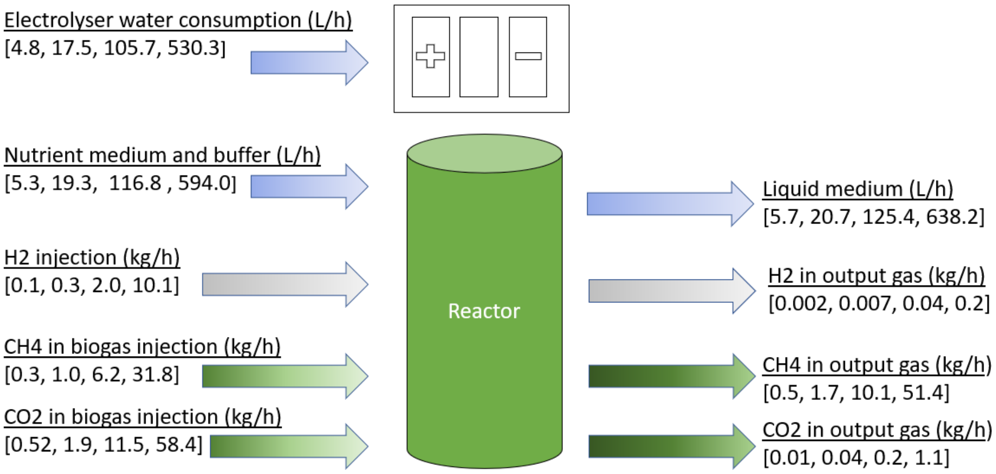

Figure 9: Illustrated representation of the mass balance of the system for all four cases. Data is represented for each case is represented: [4.8 kWe, 20 kWe, 100 kWe, 550 kWe].

4.2. Production costs

The levelized cost of CH_4 production (LCOCH4) and specific investment costs (COCH4) is shown in figure 10 for all four cases. The costs are based on both total CH_4 production including injected CH_4 ($LCOCH4_{Total}$, $COCH4_{Total}$) and CH_4 produced by BHM excluding injected CH_4 ($LCOCH4_{Meth}$, $COCH4_{Meth}$). Meaning, $LCOCH4_{Total}$ can be considered as the costs of upgrading and production of biogas while $LCOCH4_{Meth}$ is the costs of CH_4 attributed to BHM. The presented costs do not include heat recovery.

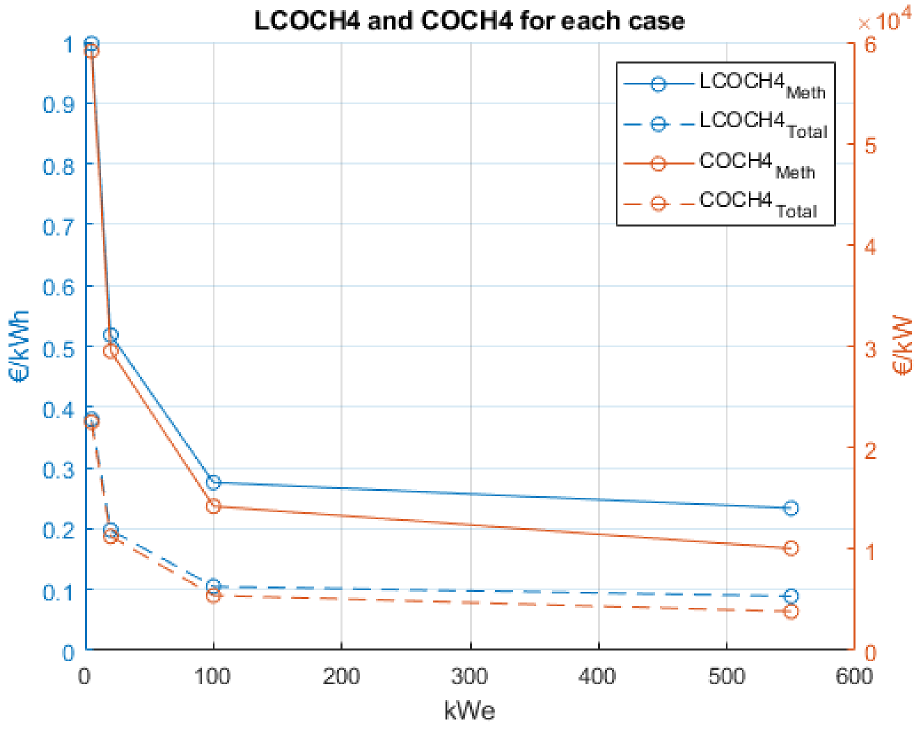

Figure 10: Resulting production costs of CH_4 and specific CAPEX for each case. $LCOCH4_{Total}$ and $COCH4_{Total}$ are based on total CH_4 production. $LCOCH4_{Meth}$ and $COCH4_{Meth}$ are based on CH_4 production from the BHM process.

An expected concept of scale is present, where the most cost-effective case is the 550-kW plant. A drop of 50% of LCOCH4 can be observed between Case 4.8 kWe and Case 20 kWe. This can be explained by the fact that at lower scales installing and most equipment costs are similar, which is shown in table 6. A less pronounced difference can be observed between 100 kWe and 550 kWe systems. This curve of scale is affected more pronounced between 550 kWe and 4.8 kWe since these cases use two electrolysers at the scale of 225 kW and 2.4 kW, respectively. While the other two cases use only 1 electrolyser at the necessary capacity, this creates a higher difference of costs between 4.8 to 20 and a lower difference between 100 and 550.

Comparing investment costs to literature is difficult due to the lack of data for smaller scales of BHM plants. However, a comparison between the result and estimated investment costs from calculations based on the studies previously mentioned in 2.5.2 can be made. Figure 11 details LCOCH4$_{meth}$ with and without electrolyser replacement. To compare, a simulation was run with a calculated total CAPEX based on equation 1 with a scale factor of 0.52 and a standardized investment cost of 600 €/kW$_{SNG}$ at 5 MW. The figure is used to compare current study's estimated total CAPEX to total CAPEX based on literature (Zauner et al. 2019).

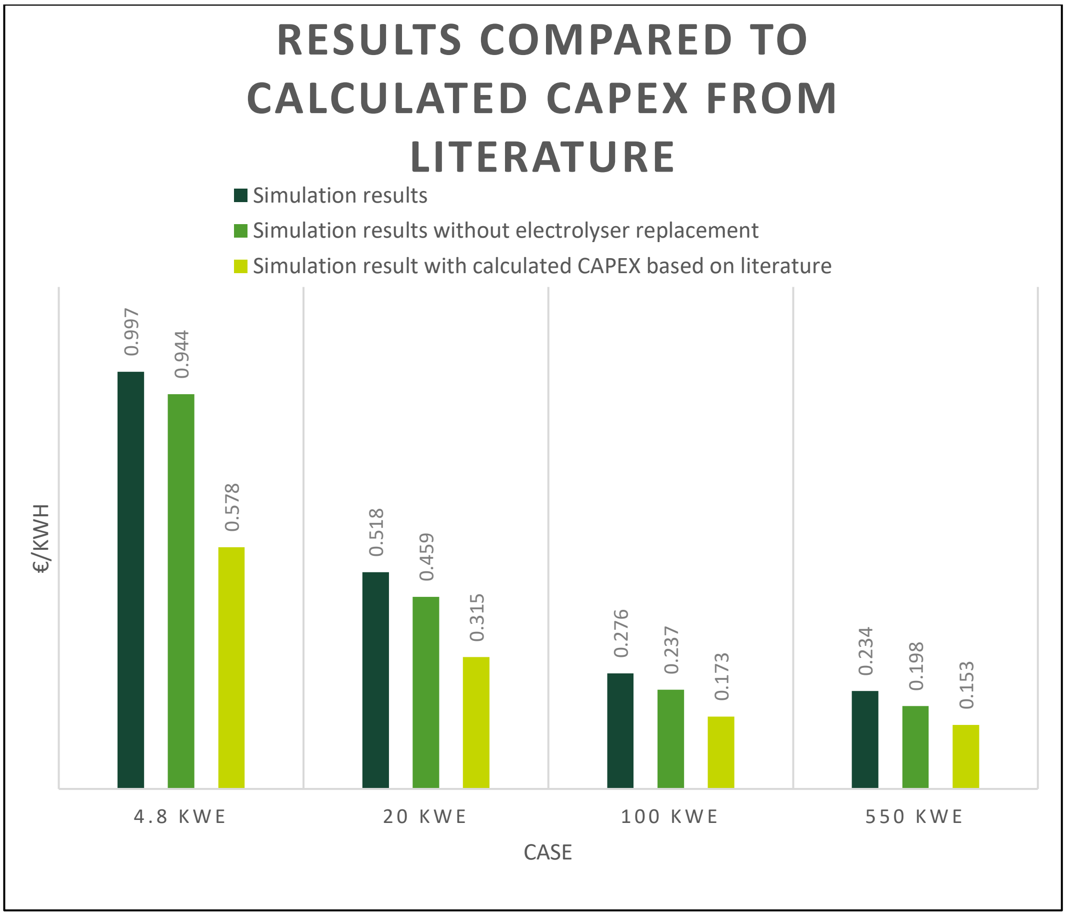

Figure 11:System LCOCH4$_{Meth}$ compared to simulations with calculated CAPEX costs based on literature for all cases.

It can be noted that base simulations result in higher production costs when compared to literature. One explanation to this is the limitation of equation 1 when comparing 5 MW scale to 5-550 kW scales. Other explanations are the high costs of electrolyser in the system. While large-scale systems of 1 to 10 MW have electrolyser costs of 500 to 1000 €/kW (Electrochaea 2014; McDonagh 2018) the current study have more than double these costs. However, this is not unreasonable due to the difference in scale. Comparison between the current study's scale and systems up to a MW scale may be misleading, resulting in difficulty to determine if the current study's results are reasonable or not.

4.3. Dependencies of the electrolyser

Both CAPEX and OPEX of the system are dependent on the electrolyser. In figure 12 the factors of OPEX are shown during one year of operation. The figure details the costs of electrolyser electricity, maintenance, staff costs and other OPEX. This includes chemicals, water, equipment electricity and purifier OPEX. It can be noted that the larger the system is, the electrolyser will have a higher impact on OPEX. Staff costs are the same for all cases (30 €/h, 207 hours) which is why it increases with lower scales.

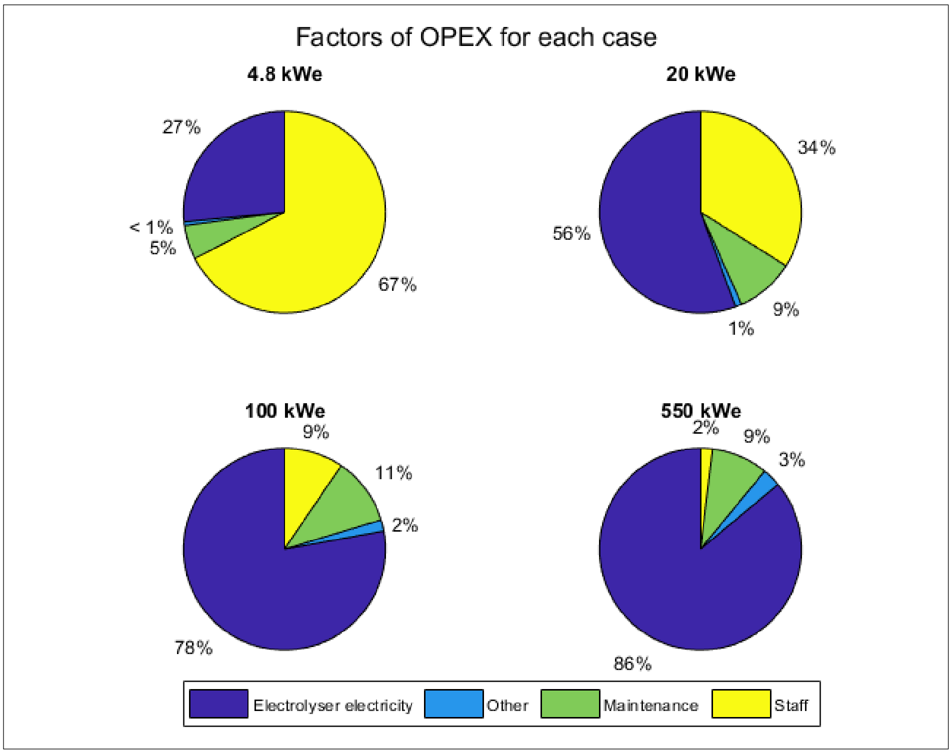

Figure 12: Overview of OPEX during one year of operation. Electrolyser electricity, maintenance, staff costs and other are shown. Other includes expenses from chemicals, water, equipment, and electricity.

Figure 13 shows the fractions of the total CAPEX. Similar to OPEX, the impact of the electrolyser grows larger as the system grows in scale. This is also evident from other studies mentioned in 2.5.2 where an increase in plant size follows an increase in electrolyser influence in costs (Electrochaea 2014).

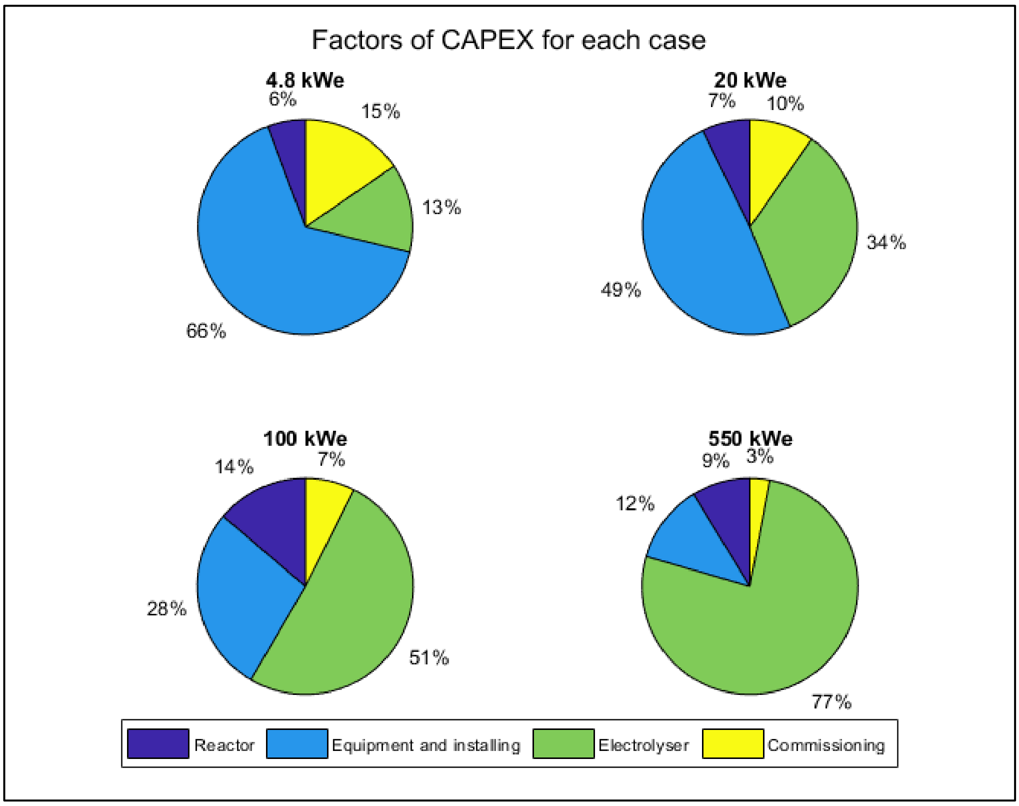

Figure 13:Overview of CAPEX for the system. Costs of electrolyser replacement are not included. The CAPEX factors are reactor, equipment, commissioning, and electrolyser.

4.4. Potential heat recovery

As can be seen in the energy balance (figure 8), some heat generated from both electrolyser and reactor is recovered. However, as previously stated in 2.1 farms using CHP will most likely not require extra heating, as 29% of the energy generated is not used or sold. If this system is used to upgrade biogas, it could be incentivized to use output gas for fuel instead of CHP. In figure 14 the $LCOCH4_{Total}$ from both base simulations and simulations using recovered heat. Cost reductions from heat recovery are assumed to substituting 0.04 €/kWh. It can be noted that a reduction is evident but does not have a significant impact on the production costs and may not be enough for a producer to turn away from CHP. This means that it could be more beneficial for the case of 20 kWe, to stack higher efficiency electrolysers that do not support heat recovery from supplier 1.

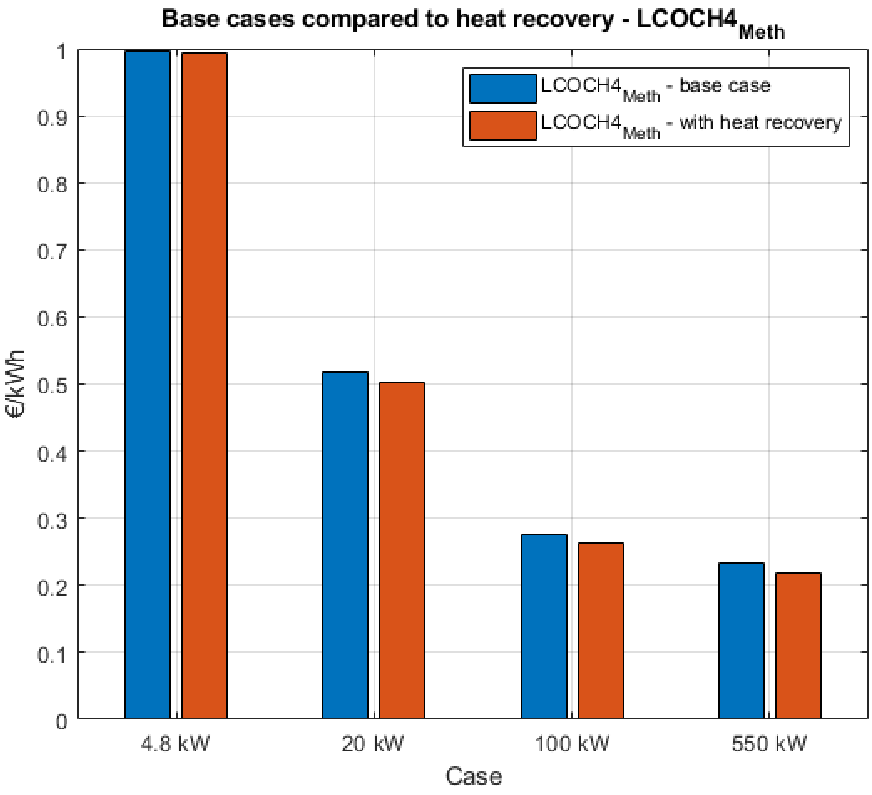

Figure 14:Resulting LCOCH4$_{Meth}$ reduction by heat recovery. Base simulation results (blue) are compared to simulation results using heat recovery (orange). Heat recovered is assumed to substitute 0.04 €/kWh of heat.

4.5. Sensitivity analysis

CAPEX

The results of the sensitivity analysis of CAPEX are shown in figure 15. The bar diagram shows the result of LCOCH4$_{Meth}$ where total CAPEX is reduced between 35%, 50% and 82.5%, and the electrolyser CAPEX is reduced by 28% and 45%. Total CAPEX reduction of 35% shows the expected cost reduction of BHM plants in 2030 (orange), 50% shows the result of a subsidy of 50% from Climate Leap (yellow) and 82.5% reduction shows the effects of both (purple). An electrolyser CAPEX reduction of 28% and 45% is the expected cost reduction of AEL and PEM electrolysers in 2030, respectively. The figure shows the impact of electrolyser CAPEX reductions depends on scale. For smaller scaled systems, the electrolyser CAPEX reduction has a smaller impact on LCOCH4$_{Total}$ than for a larger system. For case 550 kWe, a reduction of 45% of electrolyser CAPEX almost reaches the same impact as for a total CAPEX reduction of 50%. It can also be noted that the impact of total CAPEX reduction is larger for smaller scaled systems. In conclusion, larger scales of the system benefit more from electrolyser CAPEX reductions, while smaller scales of the system benefits from total CAPEX reductions such from subsidies and reductions in BHM related costs.

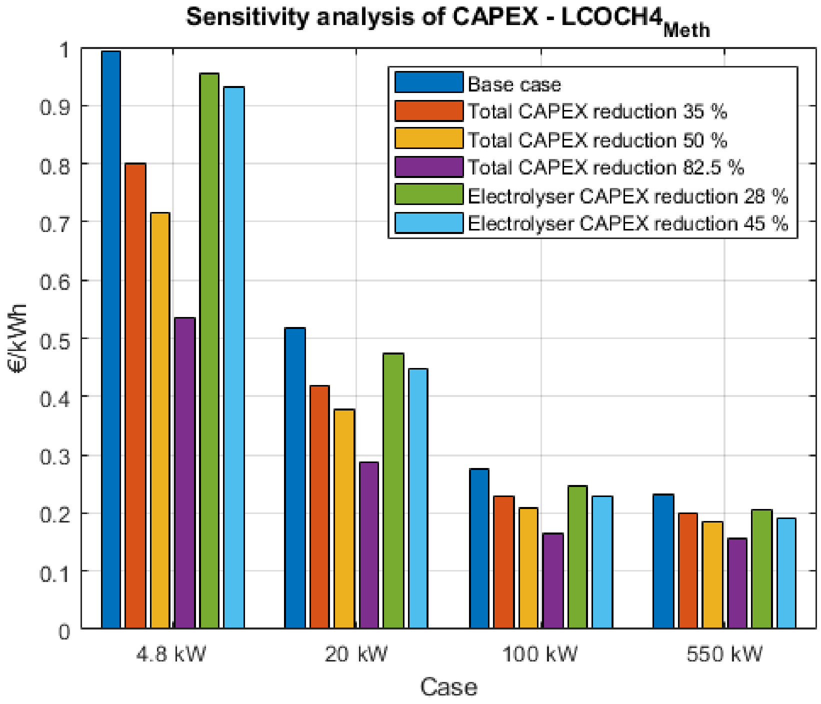

Figure 15:Sensitivity analysis of investment costs. The different CAPEX reductions are shown in the bar diagram, as well as the base case of the system with no reductions.

Electricity price

The result of the sensitivity analysis of varying electricity price is shown in figure 16. The resulting impact is the systems upgrading costs (LCOCH4$_{Total}$). The price of electricity in the simulation is set to 0.059 €/kWh which is an estimation of a static electric price. The average spot price in the year 2020 for region SE3 is 0.0221 €/kWh (Nordpool 2020). Applying this price to the simulation (60% electricity price reduction) results in a reduced LCOCH4$_{Total}$ by 20% for the largest scale. With the increase of scale, the more impact the electricity price will have on the LCOCH4. This creates a large potential for cost decrease in production/upgrading for the larger scales of the system by reducing electricity costs. Because of the high impact of the electricity price, it could be motivated to add renewable electricity sources to this system to reduce the average price.

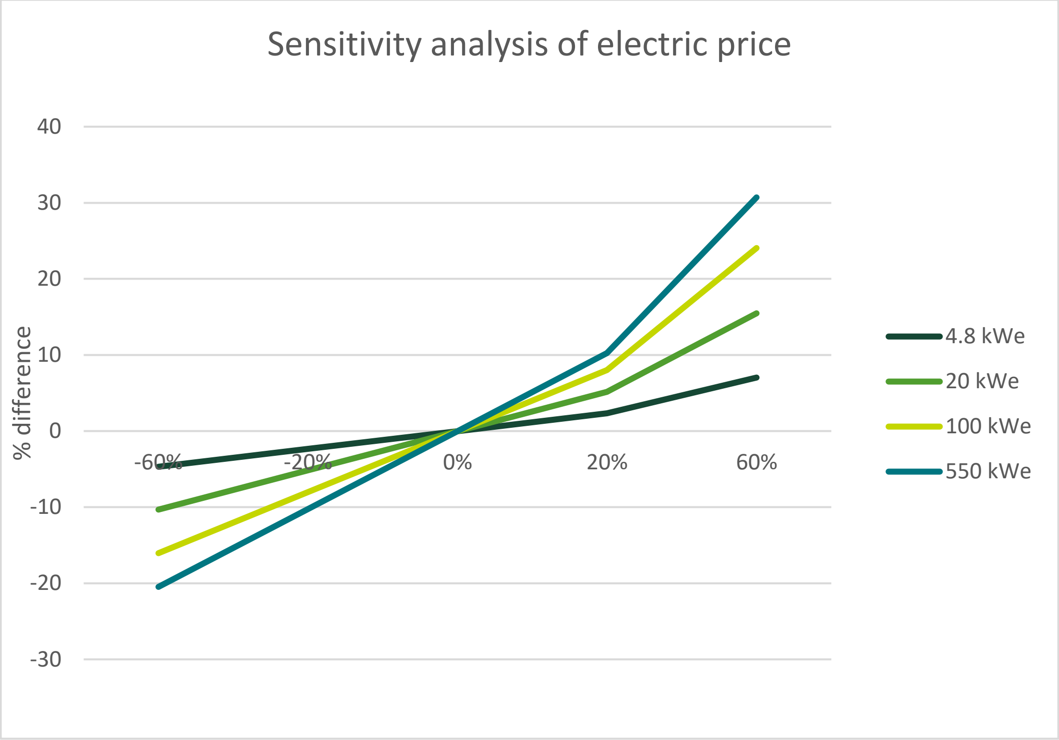

Figure 16: Resulting difference in LCOCH4$_{Total}$ from a varying electricity price. X-axis shows the variance of electricity price (%). Y-axis show the difference in resulting LCOCH4$_{Total}$ (%).

If the electricity would be available at no costs the LCOCH4$_{\text{Meth}}$ would decrease from 0.234 €/kWh to 0.114 €/kWh (decrease of 49%) for the 550 kWe system. Comparing this to results reported by a 10 MWe system, a similar decrease from 124 MWh to 55 MWh (decrease of 44%) (McDonagh et al. 2018).

Operating hours and electrolyser lifetime

The sensitivity analysis of FLH and electrolyser lifetime is presented in table 8. Only case 550 kWe is analysed with results based on LCOCH4$_{\text{Total}}$. FLH has a large impact on the resulting LCOCH4$_{\text{Total}}$. Production costs increase by 11.5% at 6800 operation hours. This simulation shows that it is beneficial to operate the system as much as possible. However, the simulation does not include electrolyser degradation which can affect the system negatively at higher FLH.

The electrolyser replacement has less impact compared to the other parameters but can still be considered important. For each replacement, the LCOCH4 increase by 7.8%. This could be a benefit if electrolysers with long lifetimes are prioritized. However, this could discourage the use of AEM. AEM is still new to the market and the effective lifetime is still unknown. Although, since the use of AEM yields low CAPEX, this could outweigh the potentially short lifetime and replacements.

Table 8: Results of sensitivity analysis of operational parameters. The effects on LCOCH4$_{Meth}$ from varying FLH, electricity price and electrolyser lifetime on case 550 kWe are investigated.

FLH					
Factor	- 50%	- 20%	-15	-10%	-5%
Result	+46.1%	+11.5%	+8.1%	+5.1%	+2.4%
Electrolyser lifetime					
Replacements	0	1	Base (2)	3	4
Result	-15.4%	-7.8%	-	+7.8%	+15.4%

4.6. Cost optimizations

The system can compete with traditional upgrading concerning performance. The system can achieve an output gas with high concentrations of CH4 and produce vehicle fuel. Another advantage of the system is its ability to increase the CH4 yield of the biogas plant, which traditional upgrading does not. While the system produces high-quality output gas, a considerable investment and operation cost are required. As observed in the sensitivity analysis, these costs can be reduced by different means.

Traditional upgrading costs for biogas plants in Sweden can be considered at 0.05 €/kWh upgraded biogas in the year 2014 for small to medium scale (Vestman et al. 2014). However, the costs change depending on the scale and at lower biogas flows the costs increase (Blom et al. 2012). Data of traditional upgrading costs in recent years were found difficult to retrieve and costs may be inaccurate for 2021.
A comparison between cost optimizations of the system and traditional upgrading costs are illustrated in figure 17. However, comparing traditional upgrading to BHM is not straightforward due to the generated CH4 in the methanation process. Therefore, the results shown in figure 16 are the upgrading costs of the system (LCOCH4$_{Total}$) with an added cost of raw biogas production from the digester estimated at 0.04 €/kWh (Vestman et al. 2014). This added cost is also added for traditional upgrading costs of 0.05 €/kWh giving a total cost of 0.09 €/kWh for produced and traditional upgraded biogas, which is illustrated in figure 17 (red dotted line).

The systems upgrading potential can also motivate the substitution of diesel. The price of diesel in 2021 is estimated at 0.12 €/kWh (excluding VAT). Cases or optimizations that have a production cost below this could motivate using BHM as upgrading to produce fuel. Figure 17 also includes the price of diesel for comparison (black dotted line).

The cost optimizations for the system shown in figure 17 are:

- B – Base cases with no optimizations.
- O1 – Heat recovery of the system is applied. Heat recovered are assumed to substitute 0.04 €/kWh.
- O2 – Total CAPEX reduction of 50% from subsidy.
- O3 – The electric price is assumed to be spot price of 2020 at 0.0221 €/kWh.
- O4 – Optimizations O1, O2 together.
- O5 – Optimizations O1, O2, O3 together.
- O6 – Optimizations O1, O2 ,O3 together in the year 2030.
- D – Diesel price in Uppsala, Sweden excluding VAT in 2021 (Dieselpriser 2021).
- TU - Traditional upgrading costs in 2014 for small to medium scale (Vestman et al. 2014).

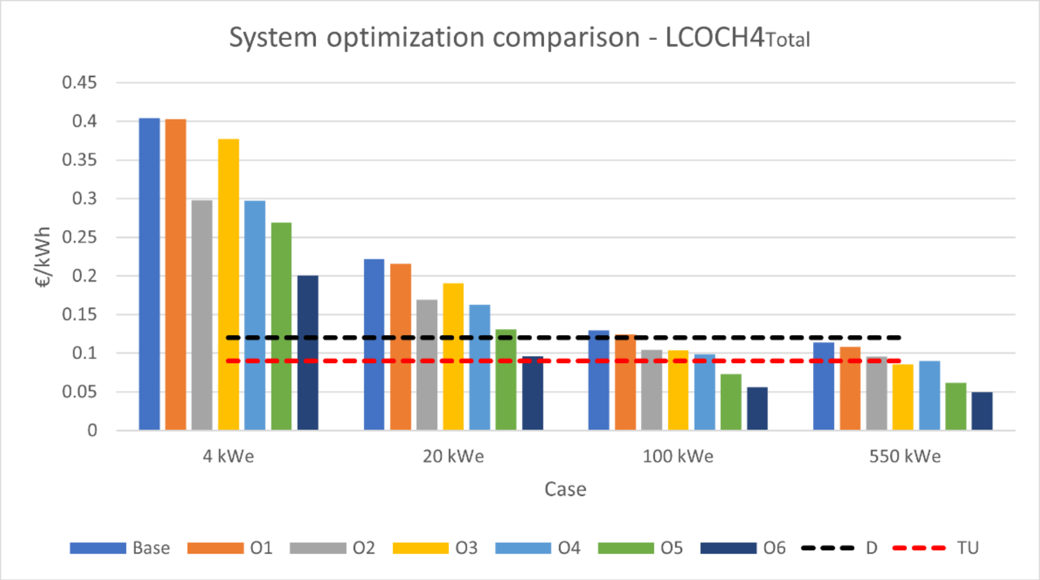

Figure 17: System upgrading costs optimizations of the system compared traditional upgrading costs and diesel price.

No base case reach economic competitiveness if compared to traditional upgrading of small- to medium-scale. The scale of 4.8 to 100 kWe systems can be considered micro scales and a direct comparison is not optimal due to traditional upgrading costs can also be expected to increase at smaller scales (Blom et al. 2012). The largest scale does reach the same or lower cost compared to traditional upgrading with all or some optimizations. By a subsidy of 50% or reducing electricity prices, this system can be considered competitive. With all optimizations, both cases of 100 kWe and 550 kWe can reach competitive upgrading costs. Both cases of 100 kWe and 550 kWe reach an upgrading cost below the cost of diesel. These systems can be motivated to be used for substituting the use of fossil fuels.

For the 4.8 and 20 kWe systems, neither reaches a competitive level of costs. It is important to remember that these systems can be considered micro-scale. As explained in 3.1.1, these cases of scale are assumed to upgrade only a part of the biogas produced. These systems could be motivated to be used for producing fuel for private use only. With all optimizations and with additional future cost reductions, the 20 kWe system can reach competitive levels in the year 2030.

This resulting comparison shows that a system at 100 kWe or higher can be a competitive option for upgrading in the future. A system at 20 kWe was shown to reach competitive levels in 2030 by utilizing all optimizations. However, these optimizations can be difficult to apply, and such a small system do need significantly more aid from subsidies to be used than at larger scales.
With further experience and efficiencies developing in the BHM market, a small-scale BHM upgrading plant can be a viable option for smaller biogas plants over 100 kWe.

As reported by McDonagh et al. 2018, secondary incomes and incentives are essential for P2G (McDonagh et al. 2018). The present study does not consider potential revenue from oxygen production or grid services which could improve upgrading costs.

4.7. Uncertainties

4.7.1. Scalability

Difficulties may arise at larger scales of the system. While electrolysers are commercially available from kW to multi-MW scales the same cannot be said for methanation reactors. No TBR in the published literature reaches the required volume for the larger scales of the system.

4.7.2. AEM lifetime

Due to no degradation of the electrolyser is assumed, the production model of H_2 supplied to the reactor may differ from a real-life plant. Lifetime cycles for AEM are still unknown and further study of AEM is necessary to know if the electrolyser type can support 8500 hours of operation for 7 years.

4.8. Future study

Future applications and studies in small-scale methanation could be done to improve the current market and development of BHM based biogas upgrading. A more thorough investment cost analysis could be done to improve the accuracy of actual costs. Grid electricity replacement such as localized renewable electricity production can be a potential addition to this system concept. Secondary incomes from oxygen can be used to further optimize the system. Further study into larger scales of TBR and higher HLR will potentially increase the reliability of the system to function at higher scales or with higher volumes of injected gases.

5. Conclusion

A concept of a biological methanation system to produce upgraded biogas at near vehicle fuel quality was created in the present study. Costs from suppliers were used to estimate electrolyser costs. Electrolysers used in the simulation is based on real commercially available electrolysers. Electrolyser types used in the four cases are AEM, AEL and PEM. The system upgrades biogas by using a TBR. The system performed competitively in upgrading performance by producing output gas with high CH4 concentration (95.7%) and H2S removal. The system also increases the CH_4 yield by 60%. However, economically the system cannot be considered competitive compared to traditional upgrading. The system, however, can be optimized at several ways to achieve higher competitiveness economically. The expected cost reduction in H2 and BHM technology in 2030 can potentially reduce the CH4 production costs by 20-25%. The concept of upgrading by BHM also reduces carbon emissions from biogas production giving the system a good chance of receiving subsidies from greenhouse gas reduction initiatives. Larger scales of the system can reach competitive upgrading costs by utilizing subsidies, electricity price reductions and heat recovery. At the lowest scale of 4.8 kWe, the systems upgrading costs are too high to motivate the use of this system at this scale. A 20 kWe system could be motivated to be used in 2030 but a considerable number of improvements and subsidies are needed to make this scale competitive to diesel prices. Scales of 100 to 550 kWe can be a competitive option for upgrading in 2021 with sufficient subsidies and optimizations. With future cost reductions and optimizations BHM could be an option for upgrading biogas in small-scale biogas plants.

References

Adnan, A. Ong, M. Nomanbhay, S. Chew, K. Show, P. (2019). *Technologies for Biogas Upgrading to Biomethane: A Review*. (ISSN 2306-5354). MPDI.

Benjaminsson, G. Benjaminsson, J. Boogh Rudberg, R. (2013). *Power to Gas – A technical review*. (SGC Rapport 2013:284) Malmö: Svenskt Gastekniskt Center AB.

Blom, H. Mccann, M. Westman, J. (2012). *Småskalig uppgradering och förädling av biogas*). Pöyry SwedPower AB.

Burkhardt, M. Busch, G. (2015). *Biocatalytic methanation of hydrogen and carbon dioxide in an anaerobic three-phase system*. **Biosource Technology**. Volume (178). 330-333. https://www.sciencedirect.com/science/article/pii/S0960852414011304

Burkhardt, M. Jordan, I. Heinrich, S. Behrens, J. Ziesche, A. Busch, G. (2019) *Long term and demand-oriented biocatalytic synthesis of highly concentrated methane in a trickle bed reactor*. **Applied Energy**. Volume (240), 818-826, ISSN 0306-2619. https://doi.org/10.1016/j.apenergy.2019.02.076

DecorexPro. (n.d.) *Isolering för rör av polyetenskum: egenskaper och egenskaper*. https://sv.decorexpro.com/uteplenie/materialy/dlya-trub-iz-vspenennogo-polietilena/ [9-8-2021]

Dieselpriser. (2021). *Dieselpriser i Uppsala.* https://bensinpriser.nu/stationer/diesel/uppsala-lan/uppsala [25-5-2021]

Electrochaea. (2014). *Final report - Power-to-Gas via Biological Catalysis (P2G-Biocat).*

Eliasson, K. (2015). *Slutrapport – Utvärdering av biogasanläggningar på gårdsnivå*. Stockholm: Hushållningssällskapens förbund.

Energigas Sverige. (2019). *Produktion och användning av biogas och rötrester år 2019*. (ER 2020:25). Bromma: Energimyndigheten. https://energimyndigheten.a-w2m.se/Home.mvc?ResourceId=179401

Engineering ToolBox, (2001). https://www.engineeringtoolbox.com [9-8-2021]

Guo, Y. Li, G. Zhou, J. Liu, Y. (2019). *Comparison between hydrogen production by alkaline water electrolysis and hydrogen production by PEM electrolysis*. **IOP Conference Series: Earth and Environmental Science.** Volume (371), Issue 4. https://doi.org/10.1088/1755-1315/371/4/042022

Jernkontorets energihandbok. (n.d). *Värmeledningsförmåga och U-värden för olika material*. https://www.energihandbok.se/konstanter/varmeledningsformaga-och-u-varden-for-olika-material [9-8-2021]

Kougias, P. Treu, L. Peñaillillo Benavente, D. Boe, K. Campanaro, S. Angelidaki, I. (2017). *Ex-situ biogas upgrading and enhancement in different reactor systems.* **Bioresource Technology.** Volume (225), 429-437, ISSN 0960-8524.https://doi.org/10.1016/j.biortech.2016.11.124

Kuckshinrichs, W. Ketelaer, T. Koj, J. (2017). *Economic Analysis of Improved Alkaline Water Electrolysis.* **Frontiers in Energy Research**. Volume (5), 1. https://www.doi.org/10.3389/fenrg.2017.00001

Lardon, L. Thorberg, D. Krosgaard, L. (2018). *Technical and economic analysis of biological methanation*. **WP3 - Biogas valorization and energy management.** D 3.2.

Lecker, B. Illi, L. Lemmer, A. Oechsner, H. (2017). *Biological hydrogen methanation – A review*. **Bioresource Technology**. Volume (245), Part A, 1220-1228, ISSN 0960-8524. https://doi.org/10.1016/j.biortech.2017.08.176

Lindorfer, J. Reiter, G. Tichler, R. Steinmüller, H. (2019). *14 - Hydrogen fuel, fuel cells, and methane*. **Managing Global Warming**, 419-453, ISBN 9780128141045. https://doi.org/10.1016/B978-0-12-814104-5.00014-4

McDonagh, S. O'Shea, R. Wall, D. Deane, J. Murphy, J. (2018). *Modelling of power-to-gas system to predict the levelised cost of energy of an advanced renewable gaseous transport fuel.* **Applied Energy**. Volume (215), 444-456, ISSN 0306-2619. https://doi.org/10.1016/j.apenergy.2018.02.019

Naturvårdsverket. (2021). *Om Klimatklivet*. https://www.naturvardsverket.se/Miljoarbete-i-samhallet/Miljoarbete-i-Sverige/Uppdelat-efter-omrade/Klimat/Om-Klimatklivet/ [24-5-2021]

Nohrstedt, L. Kristensson, J. (n.d). *Anjonubytesmembran (AEM).* [Fact sheet].

Nordpool. (2020). *Elspot Proces 2020 Monthly SEK.* https://www.nordpoolgroup.com/historical-market-data/ [25-5-2021]

Kosky, P. Balmer, R. Keat, W. Wise, G. (2013). Chapter 12 - Mechanical Engineering. Kosky, P. Balmer, R. Keat, W. Wise, G. Exploring Engineering (Third Edition). Academic Press. 259-281, ISBN 9780124158917. https://doi.org/10.1016/B978-0-12-415891-7.00012-1

Rachbauer, L. Voitl, G. Bochmann, G. Fuchs, W. (2016). *Biological biogas upgrading capacity of a hydrogenotrophic community in a trickle-bed reactor*. **Applied Energy**. Volume (180). 483-490. https://www.sciencedirect.com/science/article/pii/S0306261916310388#b0125

Rusmanis, D. O'Shea, R. Wall, D. Murphy, J. (2019). *Biological hydrogen methanation systems – an overview of design and efficiency*. **Bioengineered**, Volume (10), Issue 1, 604-634, https://doi.org/10.1080/21655979.2019.1684607

Schmidt, O. Gambhir, A. Staffell, I. Hawkes, A. Nelson, J. Few, S. (2017). *Future cost and performance of water electrolysis: An expert elicitation study*. **International Journal of Hydrogen Energy**. Volume (42), Issue (52), 30470-30492, ISSN 0360-3199. https://doi.org/10.1016/j.ijhydene.2017.10.045

Shiva Kumar, S. Himabindu, V. (2019). *Hydrogen production by PEM water electrolysis – A review.* **Materials Science for Energy Technologies.** Volume 2, Issue 3, 442-454. https://www.sciencedirect.com/science/article/pii/S2589299119300035

Sigma-Aldrich Sweden AB. (2021). *Product Comparison Guide di-Potassium hydrogen phosphate*. https://www.sigmaaldrich.com/catalog/substance/dipotassiumhydrogenphosphate17418775811411?lang=en&region=SE&gclid=CjwKCAjwv_iEBhASEiwARoemvJEm6p1JPYX3Q1Ml69k4NoKkf3NKJ7T6SWe1wcs3wUFCgYSnh51ChxoCiQoQAvD_BwE [2-6-2021]

SMHI. (2021). *Ladda ner meteorologiska observationer – Uppsala Aut, Lufttemperatur timvärde.* https://www.smhi.se/data/meteorologi/ladda-ner-meteorologiska-observationer/#param=airtemperatureInstant,stations=all,stationid=97510 [8-8-2021]

Strübing, D. Huber, B. Lebuhn, M. Drewes, J. Koch, K. (2017) *High performance* *biological methanation in a thermophilic anaerobic trickle bed reactor.* **Biosource Technology**. Volume (245). 1176-1183. https://www.sciencedirect.com/science/article/pii/S0960852417313937

Strübing, D. Moeller, A. Mößnang, B. Lebuhn, M. Drewes, J. Koch, K. (2018) *Anaerobic thermophilic trickle bed reactor as a promising technology for flexible and demand-oriented H2/CO2 biomethanation*. **Applied Energy**. Volume (232), 543-554, ISSN 0306-2619. https://doi.org/10.1016/j.apenergy.2018.09.225

Svenskt Gastekniskt Center AB (SGC). (2012). *Basdata om biogas.* http://www.sgc.se/ckfinder/userfiles/files/BasdataomBiogas2012.pdf

Ullrich, T. Lemmer, A. (2018). *Performance enhancement of biological methanation with trickle bed reactors by liquid flow modulation.* **GCB Bioenergy**, 63-71, https://doi.org/10.1111/gcbb.12547

Uppsala vatten. (2021). *Taxor dricksvatten och avloppsvatten.* https://www.uppsalavatten.se/ny-kund/taxor/taxor-dricksvatten-och-avlopp/ [2-6-2021]

Vattensystem.se (2021). *Webbutik - Silex 1 Extrapatron*. http://www.vattensystem.se/silex-1-extrapatron-2 [2-6-2021]

Vattenfall (2021). *Energiskatt på el för 2020 och 2021*. https://www.vattenfall.se/foretag/elavtal/energiskatter/ [25-5-2021]

Vestman, J. Liljemark, S. Svensson, M. (2014). *Cost benchmarking of the production and distribution of biomethane/CNG in Sweden*. Svenskt Gastekniskt Center AB. http://www.sgc.se/ckfinder/userfiles/files/SGC296_v2.pdf

Vincent, I. Bessarabov, D. (2018). *Low cost hydrogen production by anion exchange membrane electrolysis: A review*. **Renewable and Sustainable Energy Reviews.** Volume (81), Part 2, 1690-1704, ISSN 1364-0321. https://doi.org/10.1016/j.rser.2017.05.258

Appendix 1

Collected electrolyser CAPEX

Table below shows collected CAPEX costs from electrolyser suppliers. Suppliers were contacted by mail and provided ballpark figures of electrolyser price. The rated capacity, specific price and electrolyser type is noted. Electrolysers used in the current study are **marked**.

Rated Capacity (kWe)	CAPEX (€/kWe)	Electrolyser type	Supplier
62	4024	PEM	Supplier 4
120	2751	PEM	Supplier 4
175	2367	PEM	Supplier 4
225	**2363**	**PEM**	**Supplier 3**
15	7397	PEM	Supplier 4
28	4750	PEM	Supplier 4
41	3873	PEM	Supplier 4
6	9093	PEM	Supplier 4
100	3500	PEM	Supplier 6
2.5	5598	AEL	Supplier 5
2.4	**3600**	**AEM**	**Supplier 1**

Appendix 2

Detailed energy and mass balance

Detailed energy and mass balance are shown in the table below.

Energy balance results.

	4.8 kW	**20 kW**	**100 kW**	**550 kW**
	Energy input (kW)			
Electrolyser rated power	4.8	20	100	550
Equipment rated power	0.006	0.02	0.13	0.8
Energy content in injected biogas	3.9	14.4	86.8	442.1
	Energy output (kW)			
Energy content in injected H_2 (HHV)	3.6	12.9	78.0	397.1
CH_4 energy content in output gas	6.4	23.2	140.5	715.0
H_2 energy content in output gas	0.07	0.3	1.6	7.9
Heat generated from methanation	0.51	1.8	11.2	56.8
Heat transmission losses	0.09	0.2	0.4	1.3
Heat losses from heating liquid and gas	0.3	1.0	5.8	29.4
Heat generated from electrolyser	0.8	3.4	17.0	93.5
Heat recovered	0.16	3.7	18.7	101.0
	Liquid input (L/h)			
Total water usage (L/h)	5.6	20.0	121.7	700.3
Nutrient and buffer input (L/h)	4.8	17.5	105.7	530.3
H_2 input (kg/h)	0.09	0.3	2.0	10.1
CH_4 input (kg/h)	0.3	1.0	6.3	31.8
	Liquid output (L/h)			
Nutrient and buffer output (L/h)	5.7	20.7	125.4	638.2
H_2 output (kg/h)	0.002	0.007	0.04	0.2
CH_4 output (kg/h)	0.45911	1.6712	10.1	51.42

Appendix 3

Gas, liquid and material constants

In the table below, constants for gases, liquids and materials are detailed.

Constants used in calculations

Constant	Value	Source
Activated carbon density	0.63 kg/l	Engineering ToolBox (2001)
H_2S density	1.52 kg/m^3	Engineering ToolBox (2001)
H_2 density	0.08988 kg/m^3	Engineering ToolBox (2001)
CO_2 density	1.977 kg/m^3	Engineering ToolBox (2001)
H_2O density	1000 kg/m^3	Engineering ToolBox (2001)
CH_4 LVH	9.97 kJ/kg	Engineering ToolBox (2001)
H_2 HHV	142 MJ/kg	Engineering ToolBox (2001)
H_2 specific heat capacity	14.32 kJ/kg °K	Engineering ToolBox (2001)
CH_4 specific heat capacity	2.232 kJ/kg °K	Engineering ToolBox (2001)
H_2O specific heat capacity	4.2 kJ/kg °K	Engineering ToolBox (2001)
Insulation thermal conductivity	0.04 W/m^2 °K	Decorexpro (n.d.)
Steel thermal conductivity	17 W/m^2 °K	Jernkontorets energihandbok (n.d)
Passive convection of air	5 W/m^2 °K	Kosky et al. (2013)
Passive convection of water	500 W/m^2 °K	Kosky et al. (2013)

SLU
Institutionen för energi och teknik
Box 7032
750 07 UPPSALA
Tel. 018-67 10 00
pdf.fil: www.slu.se/energiochteknik

SLU
Department of Energy and Technology
P. O. Box 7032
SE-750 07 UPPSALA
SWEDEN
Phone +46 18 671000