

Main Final Report Part 1

Introduction, Purpose and Organisation



Biogas-SOEC *Electrochemical upgrading of biogas to pipeline quality by means of SOEC electrolysis*

ForskNG 2011 Project no. 10677

Front page picture courtesy of Xergi

Table of contents

1	INTRODUCTION.....	3
2	ORGANISATION.....	5
3	PROJECT DESCRIPTION	5

1 Introduction

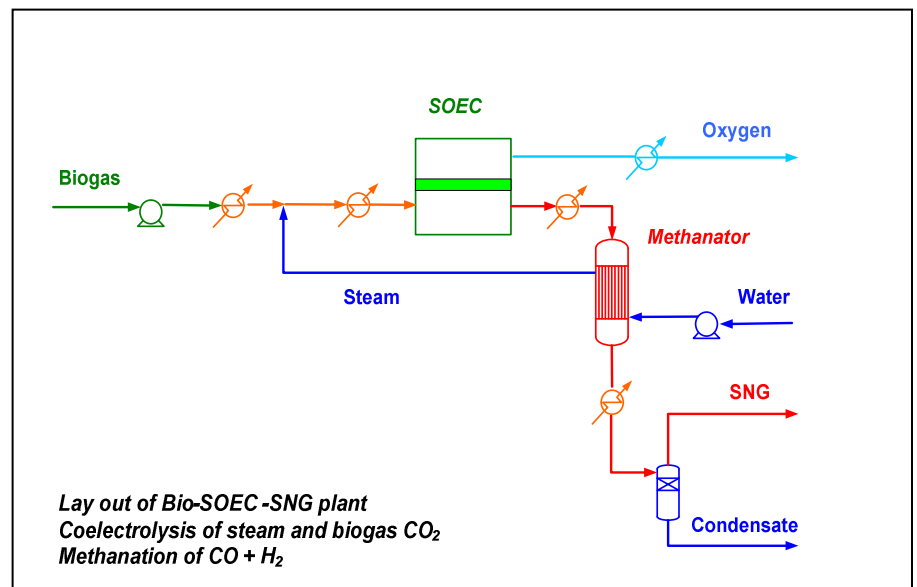
The purpose of the project was to investigate the feasibility of electrochemical upgrading of biogas to pipeline quality in Denmark by means of Solid Oxide Electrolyser Cells (SOEC). The feasibility was investigated by means of engineering studies, energy system integration and economic scenario analyses and performance of a few critical experiments with SOEC cells. The outcome endeavoured to provide answers to the following questions:

- What is the most cost effective upgrading route: Co electrolysis of the CO₂ in the biogas with steam followed by methanation or steam electrolysis followed by methanation of CO₂ in the biogas
- Can the steam reforming activity be controlled by sulphur without sacrificing too much electrochemical activity or lifetime of the cells
- What will be the overall benefits for the Denmark and under which conditions will the technology be commercially attractive

There are political goals in Denmark to utilise 50 % of the livestock manure for biogas production by 2020 and at the same time 50 % of the electricity need should be covered by wind power. Simultaneous attainment of these goals will require extensive modification to the existing energy system. If the biogas is upgraded to pipeline quality the present constraining tie to combined heat and power production can be removed and new markets, eventually including the transport sector can be made accessible. Production of “synthetic” natural gas by upgrading biogas by means of SOEC and wind power (Biogas-SOEC) can also act to store renewable energy and may also provide various balancing services to the power grid (up and down regulation of either electricity consumption or production).

State of the art technology for biogas upgrading is based on removing the CO₂ in the biogas by washing or pressure swing adsorption. The technologies are relatively expensive and add a cost in the order of 0,8 – 0,9 Dkr. per Nm³ biogas. The separated CO₂ will also contain small amounts of methane, which eventually will act as greenhouse gas.

If instead the CO₂ in the biogas is co electrolysed with steam to produce CO and H₂ (see figure on the right), the synthesis gas can be converted to methane at pipeline quality at relatively low pressure. The present SOEC electrodes, based on nickel are, however, active for the reverse reaction of methanation: steam reforming. This will result in an inefficient plant. There is the possibility to leave a small amount of sulphur in



the biogas feed to the SOEC, which will reduce the steam reforming activity to almost zero without sacrificing too much of the electrochemical electrolysis activity. This hypothesis has been investigated experimentally.

Should this strategy prove uneconomical another plant layout as shown in the figure below could be used. Steam is electrolysed to hydrogen separately and mixed with the cleaned biogas and then the CO₂ is converted to methane. In both lay outs the steam can be raised by means of high temperature waste heat generated by the methanation reaction. The steam electrolysis route has to be carried out at higher pressures in order to meet the pipeline quality requirements. Preliminary calculations showed that both routes will have low electricity consumption for the SOEC stacks of around 13 to 14 kWh per Nm³ additional methane generated in the plant. The lower heat value of methane is 9.94 kWh/Nm³. The overall efficiency of the stacks are close to 100 % calculated on the lower heat value of CO and hydrogen generated, but the transformation into methane will be accompanied by heat generation. This heat can, however, be used to raise steam and the surplus used in the biogas plant.

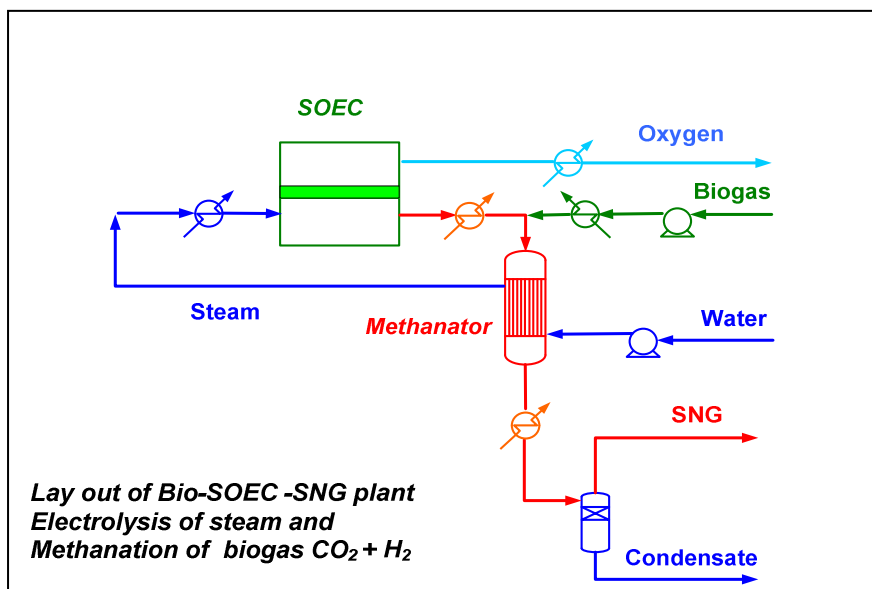
Electrolysis of steam can also be carried out using commercial alkaline or polymer based electrolysis cells but the electricity consumption will be approximately 60 and 40 % higher.

SOEC technology thus holds the potential for significantly lower cost of upgraded biogas production.

SOEC technology is still in an early R&D stage, however years of extensive R&D within SOFC

technology has been undertaken. A world leading R&D effort on SOEC is already taking place in Denmark In connection with the now commercial high temperature methanation technology developed by Haldor Topsøe A/S this provides a strong platform for an accelerated commercialisation. The project has also developed an executable R&D and commercialisation plan for the next steps to the market.

The project participants complemented each other and cover the entire chain from basic research and system development through design, marketing and sales of the Biogas-SOEC technology. The participants also represent the leading organizations in Denmark with the required competences to analyse, develop and commercialize the Biogas-SOEC technology for the domestic as well as international markets.



2 Organisation

The project has been coordinated by Haldor Topsøe A/S, which is also responsible for the system engineering studies as well as preliminary cost estimates. The main responsible has been John Bøgild Hansen.

Topsoe Fuel Cell A/S has supplied SOEC cells to the project and assessed the results. The work has been supervised by Jens Ulrik Nielsen.

Ea Energyanalysis has carried out the socio-economic analysis. Main project leads have been Felicia Fock and Hans Henrik Lindboe.

DTU Department of Energy Conversion and Storage has carried out the experimental work on the SOEC cells. Sune Ebbesen and Mogens Mogensen has been responsible for the work.

3 Project Description

The project objectives were to investigate the feasibility of electrochemical upgrading of biogas to pipeline quality in Denmark by means of Solid Oxide Electrolyser Cells (SOEC).

In principle the upgrading can be done via two different routes: A) Co electrolysis of steam and the CO₂ present in the biogas followed by a methanation step. B) Steam electrolysis, mixing the produced hydrogen into the biogas and converting the CO₂ with hydrogen in a methanation step. In the SOEC the cathode today is based on nickel cermets, as they possess the highest electrochemical activity, are easy to manufacture and cost effective. They are also used in commercial SOFC cells. These nickel cermets also have a high activity for steam reforming, e.g. the reverse process of methanation. If this reforming activity is not moderated or eliminated the effect for a plant according to route A) would be to use electricity in the SOEC to reform methane to synthesis gas, which would subsequently be converted back to methane in the methanator. The net effect would be degradation of electric power to waste heat leading to an inefficient plant.

From work on steam reforming catalyst as well as SOFC cells it is known that the steam reforming rate drops dramatically even at very low concentrations of hydrogen sulphide. The electrochemical activity is less affected at least for cells operating in SOFC mode. The idea is thus to remove hydrogen sulphide from the biogas but leave a small fraction which would control the steam reforming rate without decrease the SOEC performance unduly. The principle of controlling the steam reforming of SOEC cathodes had not been demonstrated by laboratory experiments before this project.

Steam electrolysis by SOEC has, however, been demonstrated on stack level and under pressure on the cell level. In order to meet the pipeline quality requirements a plant according to route B) would, however, have to operate at higher pressures than a plant A type.

The methanation of CO₂ is also more demanding with respect to catalyst performance and reactor volumes.

The project activities has been:

- Analyse system designs for Biogas-SOEC plants of type A and B by establishing energy- and mass balances in the form of flow sheets. This activity will provide input for the energy integration and economic scenario analysis as well as to the experimental work.
- Carry out a systematic analysis of energy system parameters and electricity market aspects and their impact on technology economy. The analysis was carried out as scenario analyses towards 2030 with the purpose of finding and describing the factors determining benefit of including Biogas-SOEC facilities in the energy system.
- Manufacture and supply state of the art SOFC cells with high sulphur tolerance for experimental tests.
- Perform critical experimental tests to determine the impact of sulphur on steam reforming rate and SOEC activity and durability and benchmark against sulphur free operation.
- Based on above issue a report on the feasibility of the Biogas-SOEC concepts, disseminate the results and develop a plan for continued R&D efforts, demonstration and commercialisation activities

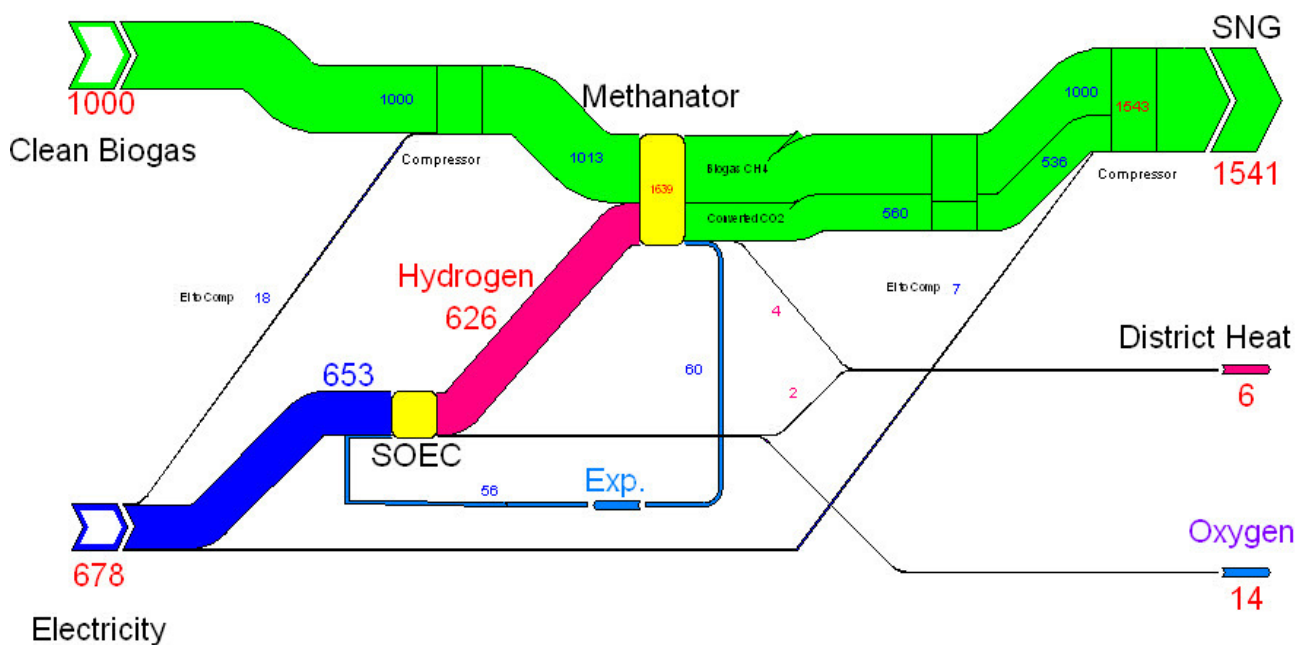
The results from this pre investigation project re documented in this report, which is divided into 6 main parts:

1. Introduction, organisation and project description
2. System and engineering studies and evaluation of SOEC performance
3. Energy system integration and economy
4. Experimental studies
5. Dissemination & continuation
6. Conclusions

Part 2

System Engineering Studies

Power to Gas Exergy Efficiency 80.2 %



Biogas-SOEC
*Electrochemical upgrading of biogas to pipeline quality
 by means of SOEC electrolysis*

ForskNG 2011 Project no. 10677

Table of contents

1	INTRODUCTION.....	3
2	PLANT CONFIGURATION A:	3
3	FLOW SHEETS AND PROCESS DESCRIPTION FOR CASE A.....	4
3.1	ENERGY CONVERSION EFFICIENCY PLANT LAYOUT A.....	6
4	FLOW SHEETS AND PROCESS DESCRIPTION FOR CASE B.....	10
4.1	ENERGY CONVERSION EFFICIENCY PLANT LAYOUT B.....	11
5	COMPARISON OF CONFIGURATION A AND B.....	12
6	PERFORMANCE EVALUATION OF SOEC CELLS DELIVERED TO DTU RISØ.....	16
7	COST ESTIMATION.....	16

1 Introduction

The two options for upgrading biogas via SOEC technology:

- A: Electrolysis of steam and subsequent methanation of the CO₂ in the biogas
- B: Co electrolysis of steam together with the CO₂ in the biogas followed by methanation of the resulting synthesis gas mixture

have been simulated by means of Haldor Topsøe proprietary computer modelling software, which is capable of calculating rigorous heat and mass balances incorporating detailed models for SOEC stacks and catalytic reactors.

The Plant capacity investigated has been nominally 7.5 million Nm³/year with a CO₂ content of 35 %. This is corresponding to approx. 50 % more than the production a typical biogas reactor installation today at f.inst. Lemvig Biogas.

Based on these calculations flow sheets have been established and the major process equipment have been sized. This has formed basis for cost estimates, which due to the novelty of the technology necessarily is of a preliminary nature, but should be accurate enough to form basis for an evaluation of the viability of the concepts.

2 Plant Configuration A:

A SOEC based biogas upgrading plant is basically consisting of four major building blocks as outlined in the block diagram on Fig. 1

- A SOEC plant producing hydrogen from steam and power from the grid
- A biogas clean up unit, which removes organic sulfur compounds, oxygen and other obnoxious compounds which would be harmful for the methanator catalyst.
- A methanation plant where the CO₂ content in the cleaned biogas is converted with hydrogen from the SOEC plant into additional methane. The strongly exothermic heat of reaction released in the methanator is used to generate steam which is used in the SOEC plant
- A final gas conditioning unit, which is removing the water content down to the level where it complies with pipeline quality and finally the gas is compressed to the required pipeline pressure

In addition to these major process blocks the residual waste heat is recuperated and used in the biogas producing plant itself or for district heating. There will also be provisions for storage of hydrogen and/or biogas due to the intermittent nature of renewable power and thus fluctuating prices.

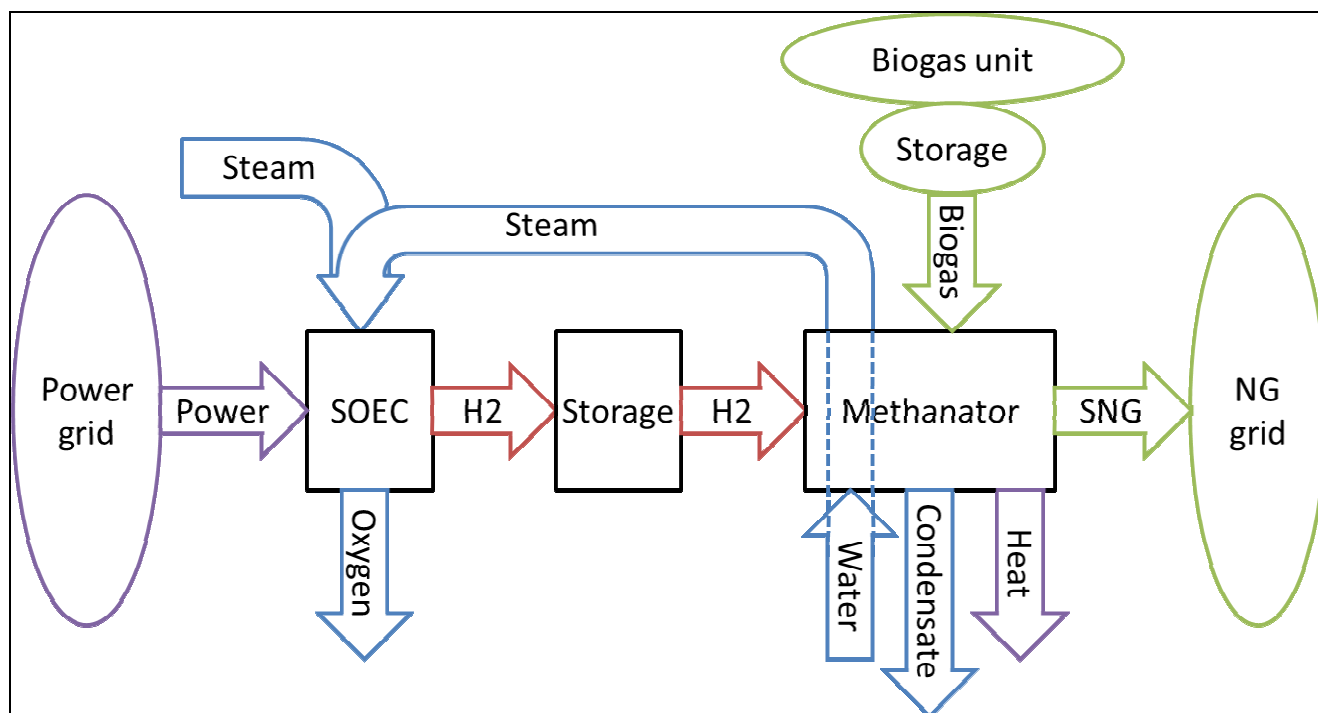


Fig. 1

3 Flow sheets and process description for Case A

The major results for the CO₂ methanation case are shown on Flowsheets 1 and 2 in Appendix 1.

Medium pressure steam from the methanation plant is preheated to the operating temperature of the SOEC around 750 °C first by feed/effluent heat exchange with the product streams from the SOEC.

This product stream consists of hydrogen, and non converted steam from the cathode, heat exchanger E205b, and oxygen from the anode, heat exchanger E205. The last temperature increase is provided by an electrical preheater, E206.

The SOEC stacks are operated at the thermoneutral voltage, e.g. there is no temperature increase or decrease across the stacks. They could have been operated in a slightly exothermic mode eliminating the need for the preheater, E 206, but as it is a cheap unit and is needed for start up purposes thermoneutral operation is preferred because it minimizes the mechanical strain on the cells.

The hydrogen and steam from the cathode is further cooled by preheating boiler feed water in heat exchanger E2040c for the steam generation in the methanator, for district heating in heat exchanger E2040b and finally by an air or water cooler E2050 before being separated into condensate – returned to the process – and hydrogen with a small water content. The cooling

of oxygen from the anode in E 207b is also used for district heating. The oxygen can potentially be used in a biological sulfur removal unit, sold or discarded.

The SOEC plant is operated at a pressure slightly above that of the methanation plant, where the operating pressure is determined by the need to obtain the correct methane content, Wobbe index etc. for pipeline SNG.

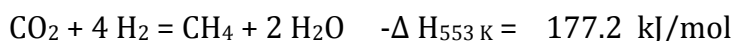
The conversion of steam at the cathode is chosen to be 80 %. Sensitivity analyses have shown that it can not go below 74 % because that would require additional electrical input to the plant to reevaporate water, in other words there is not enough heat available from the methanator alone to generate the necessary steam for the SOEC. Steam conversions above 80 % will require an undue increase in stack area.

Biogas normally contains several thousand ppm's of sulfur compounds which can be removed by biological using bacteria capable of transforming hydrogen sulfide, H₂S, into free sulfur or sulfates by reaction with oxygen from air. This will result in an unacceptable high content of nitrogen in the upgraded biogas, but if oxygen from the SOEC is used instead this problem can be eliminated.

The sulfur content can in this way be brought down to 50 – 100 ppm but even this content will be very harmful for the nickel based methanation catalyst.

After compression in a three step compressor, K1, to synthesis pressure around 16 bar g the biogas is therefore further cleaned after the biological sulfur removal by passing a train of reactors, designed to provide the optimum economical solution. It consist of one relatively inexpensive absorption masses, which removes the bulk of the sulfur, followed by another catalyst designed to remove more refractory compounds – both placed in R 100, and finally a third polisher reactor, R 200, designed to remove the last impurity traces as well as converting remaining oxygen to water by the hydrogen added from the SOEC plant.

The cleaned gas then enters the methanation reactor, R 300, where the CO₂ is converted to methane according to the reaction scheme:



The reactor is a boiling water type operating around 280 °C and the heat generated by the methanation reactor is used to generate medium pressure steam at 60 bar g.

The effluent from the methanator is used for feed/effluent preheat in heat exchanger E 200, district heating in heat exchanger E 208 b and is finally cooled to ambient temperature in the cooler E 210. The condensate is separated out and returned to the boiler feed water system. The product SNG containing at least 96 mole % methane is dried to a dew point of less than -8 °C and compressed to the required pipeline pressure of around 40 bar g in compressor K2.

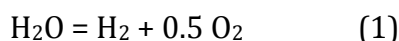
The boiler feed water is preheated in the inter stage coolers for the compressors K1 and K2 and further in BFW preheater E2040 c. The temperature of this exchanger is adjusted so that the steam production in the methanator matches the need for hydrogen production in the SOEC plant.

For the compressors a polytropic efficiency of 85 % has been used and a mechanical loss of 5 %.

The heat exchanger network has been with the appropriate constraints been optimized with a minimum pich temperature of 20 °C.

3.1 Energy conversion efficiency Plant Layout A

The by far largest energy consumption in the process takes place in the SOEC stacks. The minimum energy required to split steam into H₂ and O₂



is relatively independent of temperature and requires approx. 3.07 kWh per Nm³ H₂.

The energy of water splitting, ΔH, consists of two terms

$$\Delta H = -\Delta G + T \Delta S$$

Where -Δ G is the change in free energy, which has to be provided by electricity in the case of electrolysis, whereas T ΔS, where T is the absolute temperature and ΔS is the entropy change, can be provided in the form of waste heat for instance produced by the internal resistance of the electrolyser stacks.

The unique feature of the SOEC technology is the fact that the ASR (area specific resistance) matches quite well the T ΔS term so that

$$T \Delta S = \text{ASR} * I^2$$

at reasonable, commercially attractive current density above 0.6 A/cm².

This means that the SOEC advantageously can be operated at thermoneutral conditions (isothermal), where the efficiency defined as lower heating value of the hydrogen produced divided by the power consumption can be close to 100 %.

The competing alkaline and PEM based electrolysis furthermore operates with liquid water as feedstock, thus necessitating the use of electricity to evaporate the water also, which amount to another 0.5 kWh per Nm³ H₂. These low temperature electrolysis technologies also has ASR values, which results in production of excess heat at low temperatures employed and the power consumption, which is more than 80 % of the production price, is typically in the range of 4.4 to 5.3 kWh per Nm³ H₂.

The minimum energy requirements are illustrated on Fig. 2

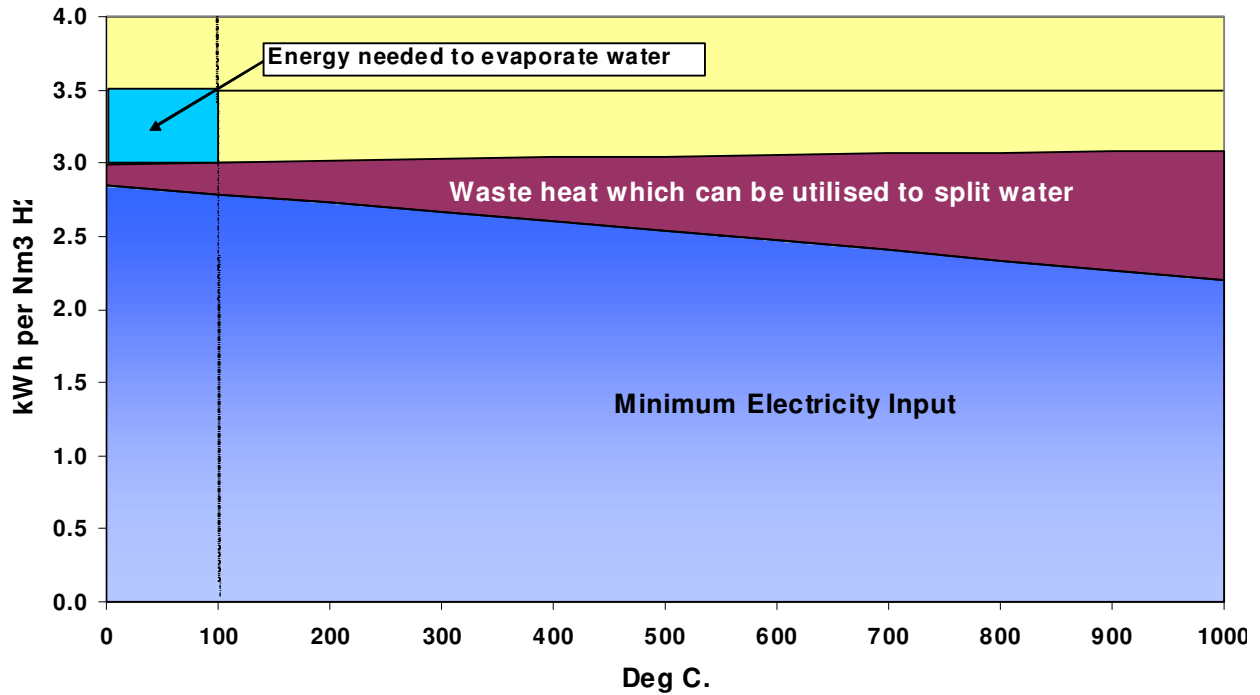


Fig. 2

Converting CO₂ to methane by laws of nature involves a loss of LHV efficiency because only part of chemical energy in the feedstock (approx. 80 %) is converted into methane whereas the rest is heat. See fig. 3.

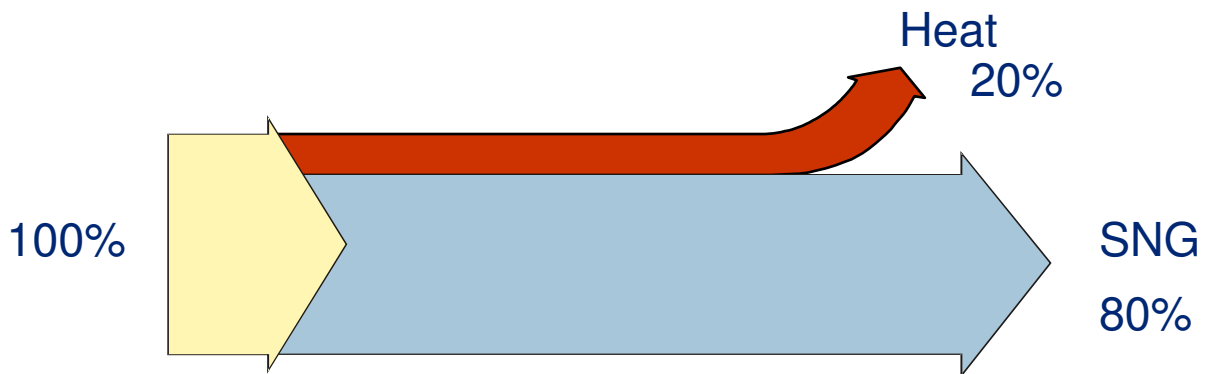


Fig. 3

The goal of the designer is then to use this heat at as high temperature as possible and in the case of biogas upgrading the heat is used directly to produce steam for the SOEC.

Haldor Topsøe A/S has been active within the field of SNG production via high temperature methanation for more than three decades and has recently obtained orders for very large scale SNG based on coal gasification in China and Korea as well as for a large demonstration plant based on wood gasification in Sweden.

Gases stemming from gasification contain only minor amounts of methane and CO₂ but has a high content of carbon monoxide. As even the best Topsøe methanation catalyst can only

withstand around 700°C for extended period of time these gases requires a lay out of several reactors in series to achieve the desired conversion as indicated on Fig. 4. It is even necessary to operate with a recycle around the first reactor to limit the temperature rise.

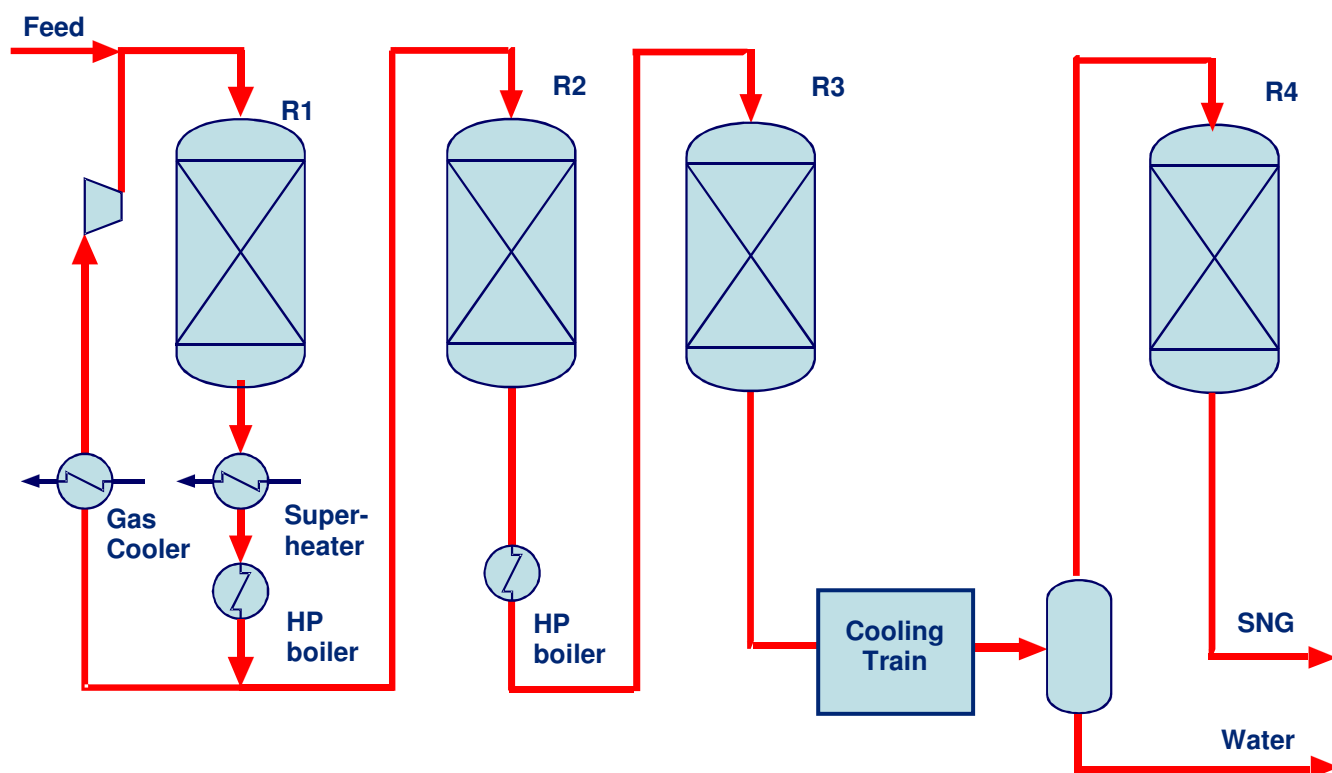


Fig. 4: Lay out of TRESP SNG plant

For the biogas upgrading case only one boiling water reactor is necessary as the gas already contains 60 – 70 % CH₄ and it is CO₂ which is methanated not CO. This makes the lay out much simpler, but although Haldor Topsøe has laboratory experience with CO₂ methanation and commercial experience with boiling water reactors for methanol, DME and gasoline synthesis the combination presented by the biogas upgrading scheme is new.

The computer calculations for Case A have found the energy conversion efficiencies shown in Table I. The electrical energy input to the process is used to drive the SOEC, the compressors and the electrical preheater before the SOEC stacks.

**Table I: Energy Conversion efficiencies
In percent of total el input**

Basis	Lower Heating Value	Exergi
SNG	76.2	80.2
District Heating	14.1	0.8
Oxygen		2.1
Total	90.3	83.0

These conversion efficiencies are remarkably high and close to the theoretical possible, but the calculation have been done with realistic compressor efficiencies, proven SOEC performance from lab scale and rigorous, pinch analyses supported heat exchanger network design with minimum pinch of 20 °C. A Sankey diagram of the exergy flows involved is shown on Fig. 5.

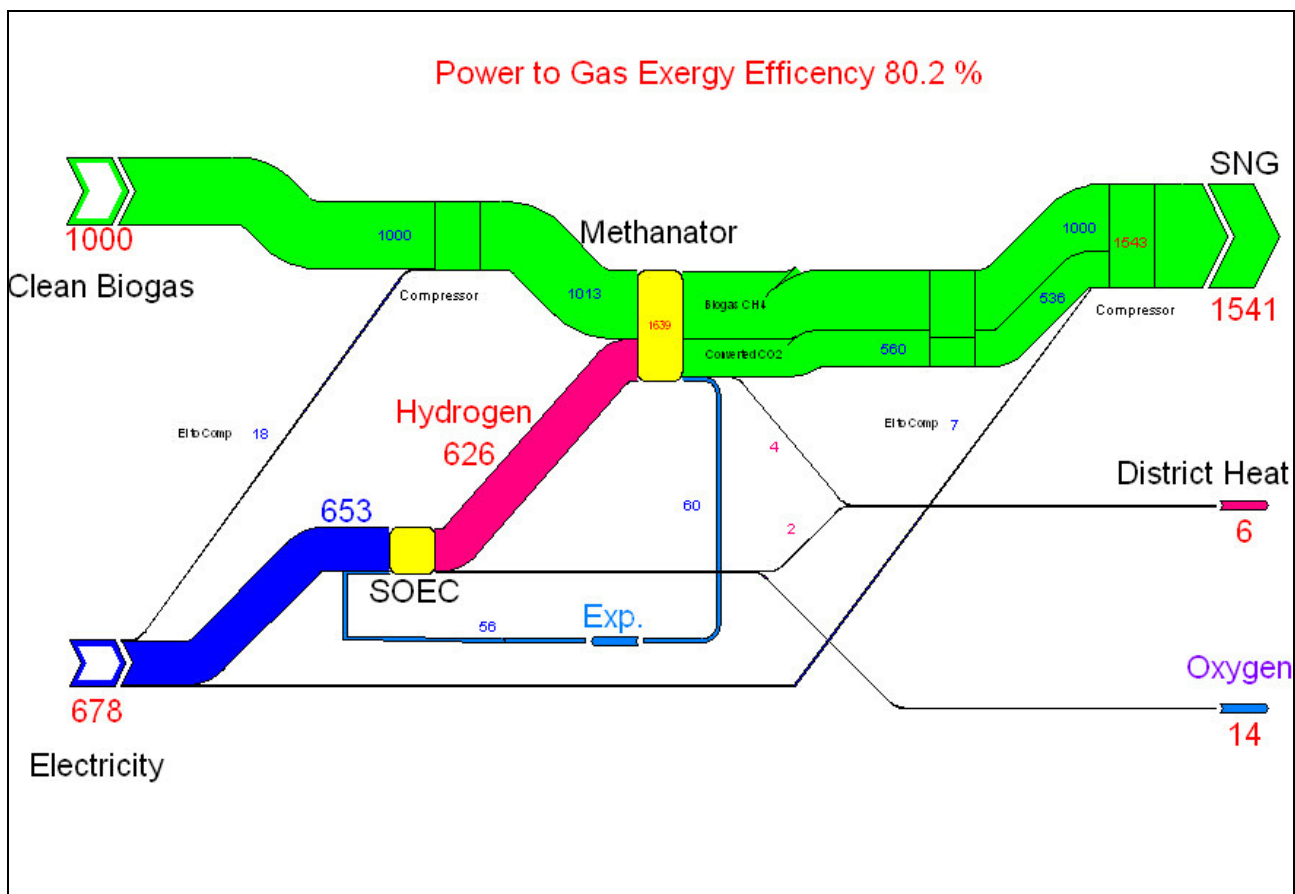


Fig. 5

As the energy content of streams in the plant consists of both chemical (LHV or HHV), latent heat, pressure and electricity it has been found most appropriate to express them in exergy

units, setting the exergy content of the methane in the incoming biogas to 1000 units. The exergy is loosely speaking the “useful” energy content of a stream, in other words the amount of energy which can be utilized to produce useful work if exchanged with the surroundings in a reference atmosphere.

The exergy concept is also very useful to analyze where the irreversible losses occurs in a process.

4 Flow sheets and process description for Case B

Compared to Case A there are a number changes to the layout because all the biogas passes through the SOEC unit but the major part of the sulphur has to be removed before. This necessitates a recycle of effluent from the SOEC in order to provide hydrogen and CO for the conversion of the oxygen in the biogas stemming from the biological biogas sulphur removal unit. The steam system is also changed in order to provide preheat for the oxygen removal reactor.

The process flow sheets for configuration B are included as Flowsheet 3-5 in Appendix 1.

After compression in a three step compressor, K1, to synthesis pressure around 9 bar g the biogas is further cleaned after the biological sulfur removal. In the first reactor, R 100, one relatively inexpensive absorption masses, which removes the bulk of the sulfur, followed by another catalyst designed to remove more refractory compounds is operating around 60 °C. Recycle gas from the SOEC unit is added and the temperature of the mixture increased to 200 °C by heat exchange with 60 bar g steam in E 110. The third polisher reactor, R 200, removes the last impurity traces as well as convert remaining oxygen to water by the hydrogen and CO added from the SOEC plant.

Medium pressure steam from the methanation plant is then mixed with the cleaned biogas and preheated to the operating temperature of the SOEC around 750 °C first by feed/effluent heat exchange with the product streams from the SOEC. This product stream consists of hydrogen, and non converted steam from the cathode, heat exchanger E205b, and oxygen from the anode, heat exchanger E205. The last temperature increase is provided by an electrical preheater, E206. The hydrogen and steam from the cathode is further cooled by preheating boiler feed water in heat exchanger E2040c for the steam generation in the methanator, for district heating in heat exchanger E2040b and finally by an air or water cooler E2050 before being separated into condensate – returned to the process – and synthesis gas plus the original biomethane with a small water content. The cooling of oxygen from the anode in E 207b is also used for district heating. The oxygen can potentially be used in a biological sulfur removal unit, sold or discarded.

The generated stream of synthesis gas together with the original biomethane is split into two streams. Around 1 % is after recompression recycled back to the gas cleaning section and the remainder sent to the methanation section where it is preheated in the feed/effluent from methanator, heat exchanger E 200. After the boiling water methanation reactor the gas is cooled in E 200 and further cooling is used for district heating in heat exchanger E 208 b and is finally cooled to ambient temperature in the cooler E 210. The condensate is separated out

and returned to the boiler feed water system. The product SNG containing at least 96 mole % methane is dried to a dew point of less than -8 °C and compressed to the required pipeline pressure of around 40 bar g in compressor K2.

The boiler feed water is preheated in the inter stage coolers for the compressors K1 and K2 and further to 275 °C in BFW preheater E2040 c. The steam production in the methanation boiling water reactor is more than required for the SOEC plant so the steam flow is split into two streams: one for the SOEC plant and the other is used to preheat biogas before the clean up reactor, R 200. The remaining heat is used for district heat production in heat exchanger, S 500.

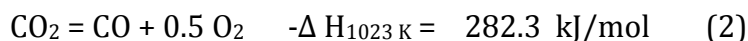
The SOEC plant is operated at a pressure slightly above that of the methanation plant, where the operating pressure is determined by the need to obtain the correct methane content, Wobbe index etc. for pipeline SNG.

4.1 Energy conversion efficiency Plant Layout B

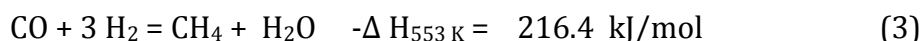
The by far largest electricity consumption in the process takes place in the SOEC stacks. In addition to the minimum energy required to split steam into H₂ and O₂



electricity is also used to split CO₂ into CO and O₂



This corresponds to 3.50 kWh for production of one Nm³ of CO from CO₂ versus 3.07 kWh for production of one Nm³ of H₂ from H₂O, e.g. 13,8 % higher energy consumption. This extra energy input is again released in the methanator because the methanation reaction:



is more exothermic than the CO₂ methanation reaction (1). Part of the extra energy input needed for CO₂ can thus be recuperated from the steam production in the methanator. The overall balances are, however, such that the heat evolution from CO methanation is more than is needed to cover the steam need for the SOEC, also because it is only required to evaporate 3 moles of water per methane mole synthesised, whereas for CO₂ methanation 4 moles of water needs to be evaporated.

The net result is unfortunately, that there a surplus production of superheated steam. In a large plant this could be utilised to generate power in an expansion turbine (of course with losses incurred compared to the SOEC electricity input), but in a relatively small biogas plant this will most likely prove uneconomical due to the high investment and low availability of such small turbines.

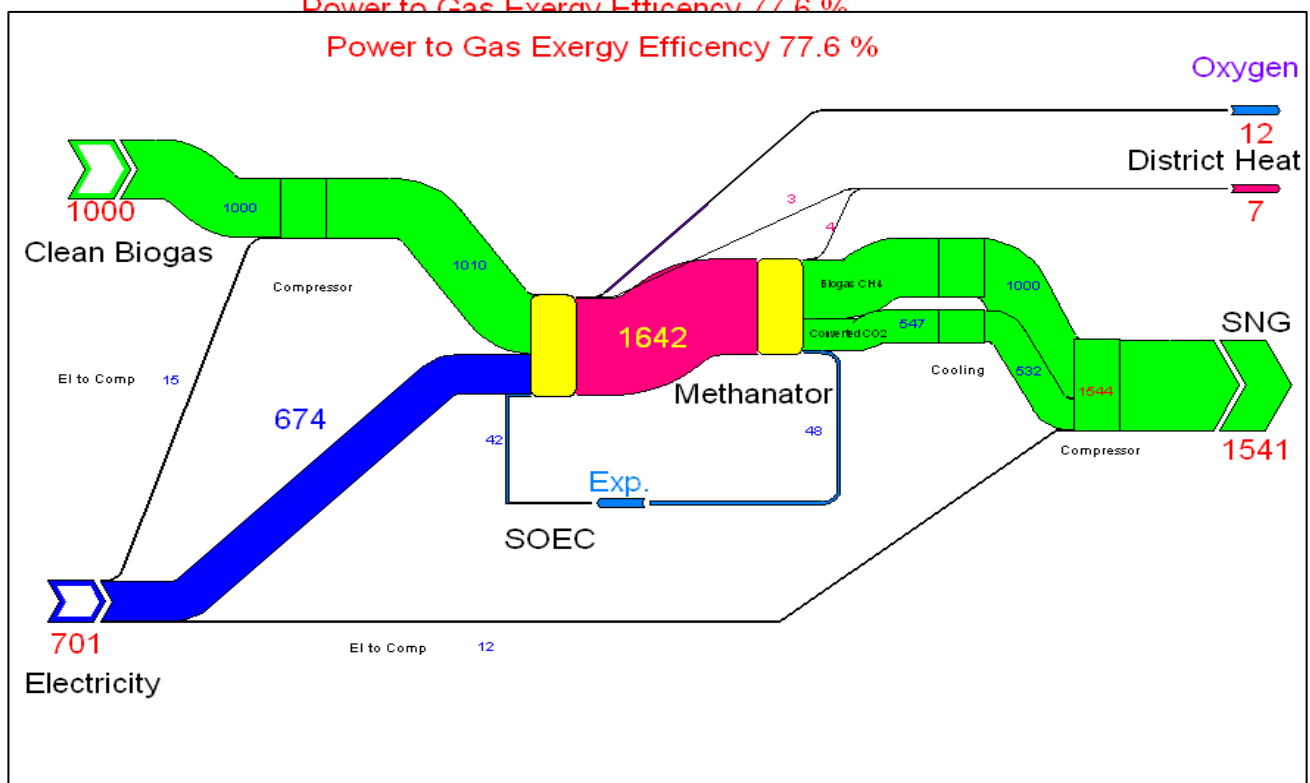
The exergy and LHV efficiency to methane is thus lower for Configuration B compared to Configuration A as it is depicted in Table 2:

**Table 2: Energy Conversion efficiencies
For Plant Configuration B
In percent of total el input**

Basis	Lower Heating Value	Exergi
SNG	73.8	77.6
District Heating	15.6	0.9
Oxygen		1.8
Total	89.4	80.3

A Sankey diagram of the exergety flows involved is shown on Fig. 6.

Power to Gas Exergy Efficiency 77.6 %
Power to Gas Exergy Efficiency 77.6 %



5 Comparison of Configuration A and B

It is interesting to compare the two configuration studied. The most pertinent data are compiled in Table 3.

Table 3 Comparison of key numbers between Configuration A and B
All units in kW except pressure in bar g

Configuration Duties in kW for	A CO ₂ Meth.	B CO Meth.	Difference B - A
SOEC	3709	3808	100
El Preheater E206	62	87	24
Comp Bio	105	85	-20
Comp SNG	40	69	28
Total el	3916	4049	133
Steam Production	595	694	100
Total DH Production	548	632	84
Biogas in exergy	5776	5776	0
SNG out exergy	8917	8918	1
P inlet, bar g	13.5	6.1	-7.4

It is observed that the electricity consumption for the SOEC is 100 kW higher in Configuration B due to the higher ΔH for CO₂ electrolysis compared to steam electrolysis as explained in section 4.1 above.

The electrical preheater, E 205, is also larger for Case B because the mass and mole flows are higher.

On the other hand the energy needed for compression of the biogas to operating pressure is 20 kW lower but as the product gas has to be compressed to pipeline pressure of 40 bar g the total compressor power for Configuration B is actually slightly higher by 8 kW. Should the upgraded biogas be used locally Configuration B would have an advantage, however.

The steam production in the boiling water reactor is 100 kW higher in Configuration B than in A, e.g. exactly the extra electrical input to the SOEC unit is recovered as heat in the methanator, but as steam at 275 °C, 60 bar g, which can not be used very efficiently in a small biogas unit as explained above.

Some of the extra latent heat manifest itself in the higher district heat production in Configuration B.

Both Configuration produce the same amount of upgraded biogas with a methane content of 96 mole % and a HHV Wobbe number of 52.0.

The operating pressure in Configuration B is 6.1 bar g inlet the methanator whereas it is 13.5 in Configuration A. This means that the equipment will be somewhat cheaper in Case B. the most important factor is, however, the reaction kinetics of the methanation reaction. Co

methanation is much faster than CO₂ methanation so the methanation section of Case B is cheaper than Case A.

The efficiencies are compared in Table 4:

Table 4
Comparison of key numbers between Configuration A and B
All units in kW except pressure in bar g

Configuration Percent efficiency	A CO ₂ Meth.	B CO Meth.	Difference B - A
Exergi eff gas	80.2	77.6	-2.6
District heating	0.8	0.9	0.1
Oxygen	2.1	1.8	-0.3
Exergy eff Total	83.1	80.3	-2.8
LHV eff gas	76.3	73.8	-2.5
LHV DH	14.0	15.6	1.6
LHV eff total	90.3	89.4	-0.9

It is seen that Configuration A is slightly more efficient than Configuration B, but that efficiency based on LHV (1st law efficiencies) are very close due to the higher amount of district heating production in case B.

The reason for the lower efficiency in case B is the net larger degradation of electrical energy to heat due to the larger ΔH for CO₂ electrolysis compared to steam electrolysis.

The more detailed breakdown of losses in the form of exergy losses are given in Table 4:

Table 4
Comparison of exergy losses between Configuration A and B

Configuration Duties in kW for	A CO₂ Meth.	B CO Meth.	Difference B - A
Total Exergy Loss, kW	760	885	124
Total Exergy Loss in %	19.0	21.5	2.5
Percent break down			
SOEC	28.80%	28.80%	0.00%
E206	2.56%	3.10%	0.54%
E205	1.55%	1.82%	0.27%
E205b	0.62%	1.14%	0.52%
E2040c	3.27%	3.38%	0.11%
E200	2.70%	2.33%	-0.37%
Methanator	20.50%	19.33%	-1.16%
Blowdown	0.14%	0.13%	-0.01%
Steam Expansion	3.00%	3.71%	0.71%
Compressors	3.78%	3.29%	-0.49%
District heating	13.22%	15.26%	2.04%
Cooling water	0.69%	0.90%	0.21%
Oxygen removal	2.17%	1.52%	-0.65%
Mixing	5.95%	5.31%	-0.65%
Condensate	0.35%	0.10%	-0.25%
Oxygen	10.60%	8.02%	-2.58%

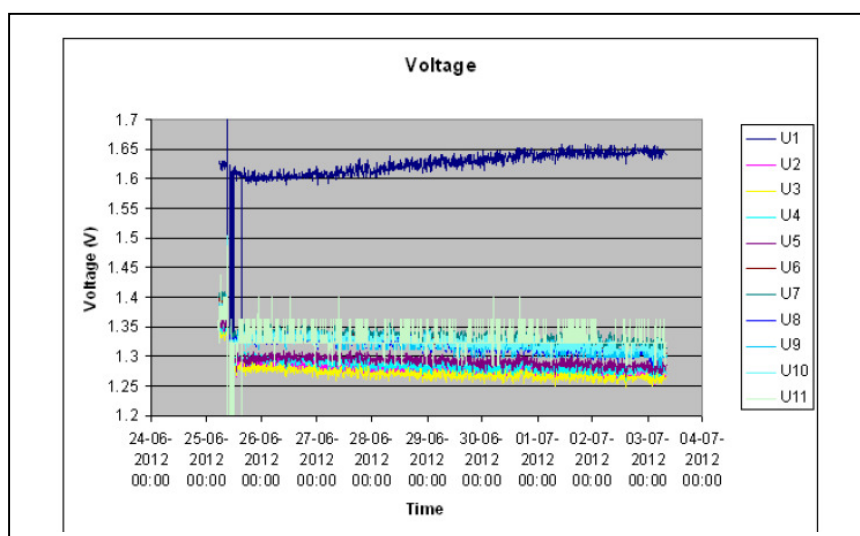
It is seen that the exergetic loss in the SOEC stacks as percent of total exergetic loss is the same in both cases.

The extra losses occur in Configuration B in the heat exchangers in the SOEC unit due to higher mass flows and especially in the steam and district heating where the extra electrical energy input is dissipated as low grade heat.

The reason why the oxygen exergy loss is lower in case B is due to the lower pressure as the flow of oxygen is identical in the two cases.

6 Performance evaluation of SOEC Cells delivered to DTU Risø

Topsoe Fuel Cell has delivered the cells tested at Risø (see Part 4 of this report) and has evaluated the performance as their contribution to Work Package 4. The initial performance has been evaluated to be in line with expectations but the degradation rate in supposedly clean gases very high and probably due to some non identified poisons in the feed gases. This is evident when comparing the results from Risø DTU in Part 4 with data obtained from a stack with 11 cells (12*12 cm²) operated with steam electrolysis at -0.6 A7cm² at Topsoe Fuel Cell.



Apart from the bottom cell, U1, which is atypical due to some contacting problems all the other cells show no sign of degradation, in fact they display a slight activation.

7 Cost estimation

Based on the flow sheet analyses of the two configurations, heat exchangers, compressors, reactors, steam system and separators have been cost estimated using Haldor Topsoe experience. It should be noted, however, that the equipment is much smaller than is normally employed in Topsoe plants so the cost databases is less reliable than usual. The reactors are also operating somewhat outside the range of normal industrial practice so the uncertainty on the cost estimates are larger than usual for such a scoping exercise, say at least plus minus 50%. A more accurate estimate can be based on vendor quotations and experience from pilot plants.

The cost estimate that a methanation plant as shown on Flowsheet 2 for Configuration A will cost approx. 20 mio DKK. For configuration B it is estimated that the plant will be 25% cheaper, due to the lower pressure and smaller reactor volume.

For the SOEC plant a price of 1000 €/Nm³ of hydrogen (or CO) produced is estimated within the timeframe consider of 2020 and 2035. For Configuration B the price will be 25 % higher due to the impact of sulphur estimated from DTU Risøe's results and the somewhat larger heat exchangers.

This mean an investment costs of 9 mio DKK for the SOEC unit in Configuration A and 11.25 mio DKK for Configuration B.



Ea Energy Analyses

Part 3

ENERGY SYSTEM INTEGRATION AND ECONOMY

Published by:

Ea Energy Analyses
Frederiksholms Kanal 4, 3. th.
1220 Copenhagen K
Denmark
T: +45 88 70 70 83
F: +45 33 32 16 61
Email: info@eaea.dk
Web: www.eaea.dk

Contents

1	Preface.....	5
2	Summary and conclusion	6
3	Background, scope and goal.....	15
	3.1 Background.....	15
	3.2 Objective and goal	16
	3.3 Scope	17
4	Framework assumptions.....	19
	4.1 Climate and energy policies.....	19
	4.2 Development in fuel and CO ₂ quota prices	21
	4.3 Development in the electricity market.....	22
	4.4 Development of biogas in Denmark.....	24
5	Two SOEC configurations	26
	5.1 Description of components	26
	5.2 Configuration 1.....	27
	5.3 Configuration 2.....	28
	5.4 Identification of critical parameters	29
6	Description of cases.....	31
	6.1 Flows.....	31
	6.2 Investments	34
	6.3 Other financial input data	35
	6.4 Value of different flows	35
7	Modelling of a biogas-SOEC unit	41
	7.1 Optimisation model.....	41
	7.2 Modelling the Biogas-SOEC using GAMS.....	43
	7.3 Mathematical model formulation	46

Results.....	50
7.4 Configuration 1.....	51
7.5 Configuration 2.....	55
7.6 Regulating power.....	56
7.7 Sensitivity analysis.....	63
Reference list.....	73
Appendix A: Subsidies for biogas.....	74
Appendix B: Energy and CO2 taxes	76

1 Preface

This report is a part of the documentation from the ForskNG funded project “Biogas-SOEC”, ForskNG project number 10677. The project is executed by Haldor Topsøe (project leader), Topsoe Fuel Cell, RISØ DTU and Ea Energy Analyses. The project was initiated in spring 2011 and is to be completed in 2012.

In the project the viability of using SOEC technology in combination with a catalytic methanisation process is analysed. The technology has potential for efficient conversion of electricity to gas. If there are many hours of the year where the electricity market price is substantially lower than the price of green gas, the technology may be viable.

In this project the analysis focuses on using a SOEC unit and a methanator for converting biogas to pressurised SNG for direct injection in the natural gas grid.

The basic idea behind integrating a biogas/SOEC plant in the energy system is to:

- upgrade biogas to SNG for the natural gas grid
- increase the SNG production compared to normal upgrading technology by converting the excess CO₂ from the biogas and hydrogen from a SOEC unit into additional SNG
- make the SOEC unit operate (and hereby use electricity to produce hydrogen) when the electricity price is low
- use the natural gas grid as an “electricity storage”

Other applications could be interesting, including using the SOEC cells in reverse mode to produce electricity in times of scarcity. Such applications are not further analysed in this report.

This part of the final report presents the findings from Work Package 3 “Energy system integration and economy”.

2 Summary and conclusion

In this work package the costs and benefits for society of converting the CO₂ content in biogas into methane by use of SOEC-technology has been analysed for two sets of main assumptions which represent scenarios of the energy system in Denmark in the years 2020 and 2035.

Data sources

Haldor Topsoe provided estimates of input data regarding CAPEX, OPEX and conversion efficiencies of the combined SOEC and methanisation facility. Ea Energy analyses have provided price estimates on all input/output energy flows in 2020 and 2035. These estimates are derived from scenario analyses of the energy system, based on assumptions from the International Energy Agency, the Danish climate commission and own calculations.

SOEC and methanisation

In the solid oxide electrolyser cell H₂O in the form of steam is split into hydrogen and oxygen or carbon dioxide is split into carbon monoxide and oxygen. Both processes consume power. One of the advantages of the SOEC compared with traditional electrolysis is the high conversion efficiency due to the relatively high temperature (700-800 °C).

In the methanisation process the product gas from the SOEC is synthesized with CO₂ in a catalytic process to produce methane. The methanator produces excess heat.

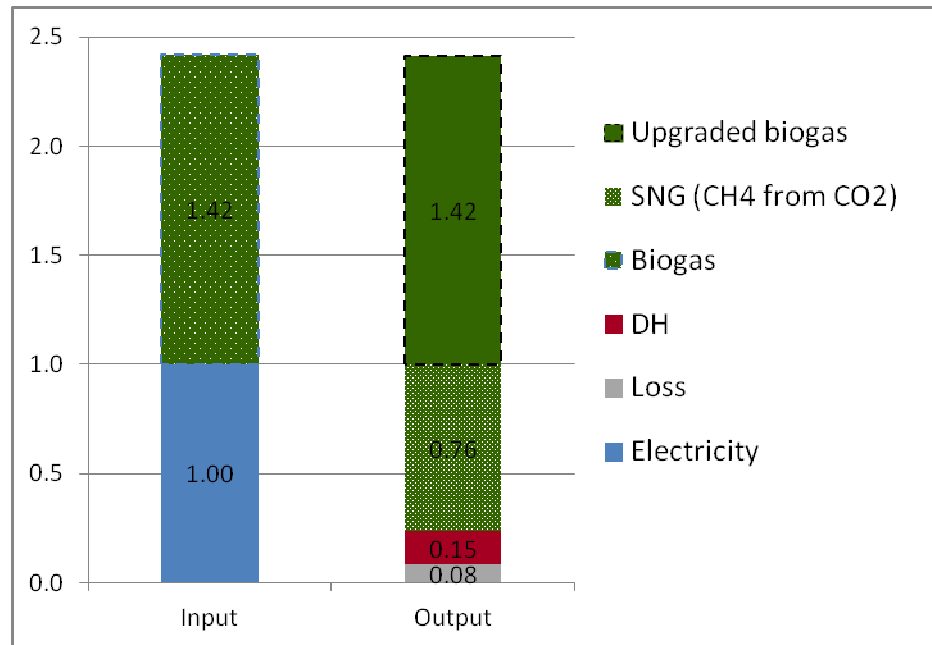


Figure 1: Energy flows in calculated standard configuration (total of the SOEC and the methanator. SNG = Substitute Natural Gas)

Figure 1 shows the energy flows in the basic SOEC and methanator configuration. All energy flows in the figure are relative to the electricity consumption (1 unit). The technical analyses in this project indicate that the plant (SOEC + methanator) is able to produce 0,76 unit substitute natural gas (CH₄) based on 1,0 unit electricity. In addition to this 0,15 units of heat can be recovered. Energy losses amount to only 0,08 units of electricity consumed.

Socio economy

Biogas consists mainly of CH₄ (65%) and CO₂ (35%) and the purpose of the SOEC application is to transform the CO₂ to methane, thus producing a gas with almost 100% CH₄ content.

The value of this transformation process comes from four sources:

- Value of substitute methane suited for direct injection into the existing natural gas system.
- Avoiding alternative cost of upgrading the biogas when necessary.
- Value of heat for district heating
- Value of regulation resources in the electricity system

Biogas can be used directly as it is or it can be upgraded to natural gas standards. It is an assumption in the economy calculations in this report that there is a future demand for upgraded pressurized biogas.

The heat production from the methanisation will partly be used to heat water for production of steam for the SOEC. Excess heat from the overall SOEC and methanisation processes that is not lost will be used for district heating or process heat at the biogas plant.

The three cost elements associated with the SOEC/methanisation process are capital costs, operational costs and most importantly, the cost of the electricity consumption.

Key data for the analysis are shown in Table 1 below:

		2035
CAPEX (8500 hrs, 5%)	DKK/GJ CH ₄ produced	41
OPEX	DKK/GJ CH ₄ produced	16
Biogas	DKK/GJ biogas input	132
Upgrading of biogas	DKK/GJ biogas upgraded	25
Heat	DKK/GJ sold	75
Natural gas	DKK/GJ	77
CO ₂ price	DKK/ton	252
SNG	DKK/GJ (upper value)	157
SNG	DKK/GJ (lower value)	91
Electricity (average)	DKK/MWh	373

Table 1: Value and cost elements of the combined SOEC and methanator used in the calculations in year 2035.

Value of methane from CO₂ (SNG)

The SNG lower value in Table 1 is based on the displacement of natural gas including saving CO₂. The upper value is based on the SNG displacing upgraded biogas, which has a significantly higher value. The value of upgraded biogas is described in "Biogas - analyse og overblik, Ea Energianalyse, 2012".

In a society moving away from the use of fossil fuels SNG will in practice displace biomass-based gases, e.g. biogas and gasification gas. Therefore the upper value of SNG will have more weight in the conclusions from this project.

Value of electricity

The average value of electricity shown in Table 1 is based on hourly calculations in the Balmorel model representing prices in a wholesale market. The price does not include taxes or tariffs for using the grid. Also "PSO" tariffs to support the transformation to renewable energy are not included. The calculations were made in 2010 for the Danish climate commission. It could

be debated to which extent PSO- and other tariffs should be included in socioeconomic electricity prices. It is probably fair to say, that if the electricity consumer is able to avoid consumption of electricity at times when the grid use is peaking, the full grid tariffs should not apply in a socioeconomic calculation.

Simple analysis

Figure 2 shows the result of a simple analysis based on the figures in Table 1. When using the high SNG value the facility shows a socioeconomic surplus. With the lower SNG value there is a deficit. Naturally, on the income side the value of SNG is the most important element. However, also avoided costs of upgrading the original biogas is significant. On the expense side the dominating figure is the value of consumed electricity.

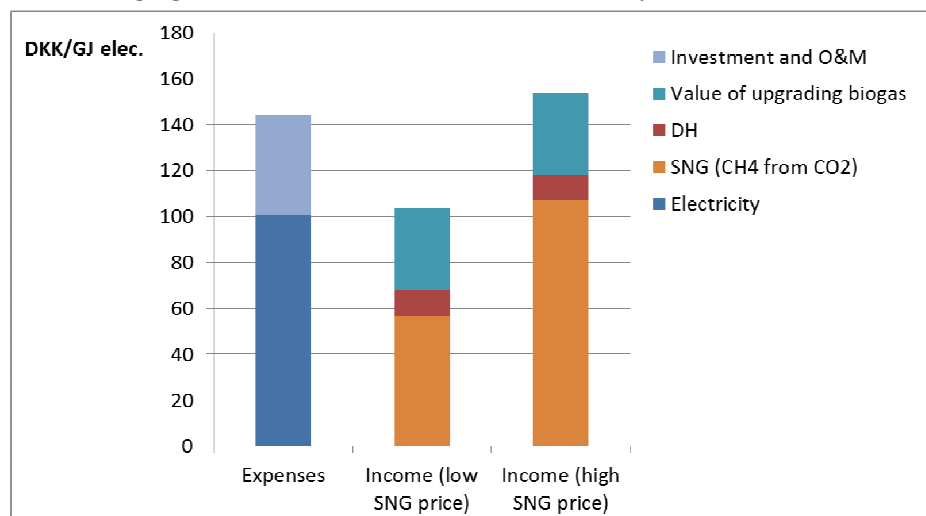


Figure 2: Simple economic analysis in 2035 based on figures in Table 1. The economy is expressed in DKK per GJ of electricity consumed in the SOEC unit.

Internal rate of return (IRR)

The ability of an investment to generate value for a number of years can be expressed as the internal rate of return. If the average electricity price is 373 DKK/MWh (calculated market price in 2035) and the high SNG value is used, the IRR of the SOEC and methanator is 22%. With an electricity price of e.g. 500 DKK/MWh the IRR is calculated to be only 3%.

The not surprising high dependence on the electricity price/SNG price ratio indicates that pursuing lower electricity prices in the market by undertaking a more flexible operational pattern than the flat 8500 hours/year used in the simple analysis could be a beneficial strategy. Especially in a future with more variable electricity prices.

Dynamic model

An optimisation model was created in GAMS for this project. The model optimises the SOEC/methanator system with respect to economy. The model

has perfect foresight of one year of hourly electricity prices (developed in the Balmorel model) in 2020 and 2035. Other model inputs are energy prices, specific investments and O&M costs.

The primary condition of the system is a fixed size of the biogas plant, and hereby a fixed hourly biogas flow. The biogas plant is assumed to be in operation 8500 h/year. The model has the possibility to invest in a SOEC unit, a methanator, a biogas storage and a hydrogen storage in order to optimise the economic performance. In the optimisation procedure the lifetime of the SOEC unit is expected to be 5 years. All other components have a 20 year lifetime.

By fixing the lifetime for 5 and 20 years respectively independent of the actual annual operating hours, dynamic operation is “punished”. This could be interpreted as a possible mechanical wear stemming from a variable operation pattern.

Two configurations

In addition to using the model for analysing the benefits of running the SOEC facility dynamically, the model is also used to evaluate two different system configurations. In the basic configuration (1) only steam is lead to the SOEC. The produced hydrogen is mixed with biogas and fed to the methanator unit. In the second configuration (2) purified biogas is led directly into the SOEC unit together with the steam. The product gases are then fed to the methanator. In configuration 2 the investment in the SOEC is higher and the investment in the methanator is lower than in configuration 1.

In configuration 1, dynamic operation of the SOEC demands investment in more SOEC cells and a hydrogen storage. In configuration 2, dynamic operation calls for investment in more SOEC cells, biogas storage AND a syngas storage (if the methanator size is unchanged). Therefore it could be expected that dynamic operation in configuration 2 will be less profitable. Main results from the analysis are seen in Table 2.

		2020	2020	2035	2035
		Low SNG	High SNG	Low SNG	High SNG
Avg. electricity market price	DKK/MWh	363	363	373	373
SNG selling price	DKK/GJ	74	140	91	57
Configuration 1					
Is dynamic operation profitable?		no	no	yes	Yes
Hours of operation for SEOC	Hours	8500	8500	7480	7480
Avg. electricity buying price	DKK/MWh	363	363	347	347
Investment year one (Mio DKK)	Mio DKK	29	29	33	33
Avg. annual profit	Mio DKK	-4,7	1,4	-3,3	2,7
Configuration 2					
Is dynamic operation profitable?		no	no	yes	Yes
Hours of operation for SEOC	Hours	8500	8500	7735	7735
Avg. electricity buying price	DKK/MWh	363	363	350	350
Investment year one (Mio DKK)	Mio DKK	27	27	32	32
Avg. annual profit	Mio DKK	-5,0	1,1	-3,7	2,5

Table 2: Results from the dynamic modelling of the SOEC and methanator units in 2020 and 2035.

In 2020 the model finds no profit from trying to optimize the operation by building gas storages and avoiding the highest electricity prices. In 2035 price fluctuations in the electricity market have increased and the model chooses to invest in more SOEC cells and gas storages. In configuration 1 the optimisation increases the SOEC investment by app 10% and adds a 7 hour hydrogen storage capacity. However, the extra SOEC cells and the storage only reduce the average electricity buying price by 7%. The relatively low effect of adding more cells indicates that the data regarding price fluctuations in the electricity market have a strong seasonal element, and are not only a day to day issue. This has not been further analysed in this project.

In configuration 2 dynamic operation is less profitable than in configuration 1 as was expected. A range of sensitivity analyses have been carried out, showing how the results change if the future investment cost of key components in a combined SOEC and methanator unit are varied.

Regulation power

The electricity prices used by the dynamic model resemble wholesale day ahead prices (spot prices). In a market like the Nordpool market, there is also a need for regulating power which is activated closer to the operating hour. The *regulating prices* represent the value (or cost) of changing your consumption away from what was originally planned. Electricity consumers,

who are active in the regulating market, can potentially buy electricity cheaper than in the spot market.

By using a Markov chain model developed in the so called *FlexPower* project, a series of regulating prices have been produced based on the 2035 spot prices used in this project. Different simple strategies for a SOEC plant to decrease the electricity cost by being active in the regulating market have been analysed.

Based on these analyses, we estimate that proper usage of regulating power prices can reduce the annual electricity payments by up to 10 %, which is a substantial figure. This is under the condition that there is full foresight of the regulating power price. Realistically the annual electricity payment can probably be reduced with up to 5 %.

This analysis has not included the technical possibility of using the SOEC cells in reverse mode, thereby actually producing power to the grid at times with very high electricity prices.

Conclusions

In this work package three main tasks have been carried out for analysing the value of SOEC technology in Denmark in 2020 and 2035. The tasks were to:

- a) Set up a set of framework conditions and price-assumptions for relevant energy flows,
- b) Project investment costs and operating costs for the main components in a future SOEC-plant and
- c) Develop and run a dynamic model for optimising the operation strategy and optimising the main components of the SOEC and methanisation units.

Benefit for society

The results show, that there could be a substantial benefit for society of deploying SOEC technology for converting the CO₂ content in biogas to CH₄. The main prerequisite for this conclusion is that the value of upgraded biogas (DKK/GJ) is higher than the value of electricity (DKK/GJ) in a sufficient amount of the hours over a year. This can be the case if:

1. Society is pursuing a goal of dramatically reducing the use of fossil fuels in a foreseeable future
2. The future electricity system is mainly based on fluctuating sources (wind power) as shown in reports from the Danish Climate

Commission, and there is a deficit of demand response in the electricity market.

3. Biomass is a scarce resource, making it costly to produce sustainable carbon based energy carriers directly from biomass.

The analysis shows surprisingly, that even when the price variations in the electricity market are substantial, the optimised SOEC plant will run more than 7000 hours/year. Sensitivity analysis show that the investment cost of the SOEC unit and the hydrogen storage must be substantially lower than in the base case before such a unit will be used with a large degree of flexibility in the market. Even if SOEC investments are 25 % lower, this would only result in reducing the share of hours with the SOEC unit in operation from 88 % to 81 %. When reducing the SOEC and the hydrogen storage investment costs with 50 %, the optimised SOEC unit will be in operation 4800 hours a year, and hereby be able mainly to run when electricity prices are low.

A basic idea behind integration of the SOEC unit into the energy system is only to use electricity when prices are low, and hereby act efficiently with demand response. However, due the necessary investments in biogas storage, hydrogen storage and, most importantly, a larger SOEC unit the demand response feature is very limited; Only the highest electricity prices are cut off.

Comparing the two process configurations

When comparing the two different process configurations (introducing the biogas before or after the SOEC unit), the analyses in this project show that the economy is quite similar, but slightly better for configuration 1 (introducing the biogas after the SOEC unit) than for configuration 2. Other parameters should be analysed further to evaluate which configuration is preferable. E.g.:

- more precise investment costs for the two configurations
- chemical pros and cons (degradation due to pollution of cells)
- analyse internal energy streams (pinch analyses)

Business case

The analyses in this report have a socio economic focus. If the future market prices on electricity and SNG will in fact be near the socio economic prices used in this report, there could be good commercial business cases in the SOEC technology. This is under the condition that it is decided that biogas must be upgraded to be used in the natural gas grid and the CO₂-SNG is seen as sustainable gas, and therefore valued as upgraded biogas.

Uncertainties regarding price prognoses

The existing public support for green gases in Denmark and other countries yields a potential selling price close to the figures used in this report. Regarding the development of prices in the electricity market, the data used should be used with more care. Major uncertainty in this respect is also how future payments for using the infrastructure in the form of net tariffs and PSO tariffs will affect a SOEC business case.

3 Background, scope and goal

3.1 Background

According to a majority of scientists and decision makers climate change and scarcity of resources are two main challenges that will face the world in the coming decades.

80-95 % GHG reduction in the EU by 2050

In February 2011 the European Council reconfirmed the objective of reducing greenhouse gas emissions by 80-95% in 2050 compared to 1990 in order keep global temperature increase below 2°C. The EU commission released “A Roadmap for moving to a competitive low carbon economy in 2050” in 2011, showing results of low-carbon scenarios for the EU. The power sector will be important in this transition where variable power sources such as wind and solar power are envisioned to play a key role.

100 % renewable energy in Denmark by 2050

In Denmark the government has a vision of an energy and transport sector using only renewable energy by 2050. The Danish Climate Commission was established by the previous government and released their final report in September 2010. In this report the Danish energy system was modelled in detail, and different fossil free scenarios were presented. In all these scenarios especially wind power, but also biomass are important contributors to the fossil free energy system in 2050.

Methane can become an important energy carrier in the future

The future role of biomass in the energy system is somewhat disputed. There have been raised concerns as to the amount of sustainable biomass resources that will be available for energy production in the future. The energy sector will compete with food, feed, aviation fuels, chemical industry etc. for the biomass resources. The world will therefore probably be searching for other energy-carriers in the form of liquids or gasses that can contribute in both the transport sector and in the energy sector.

Technologies that can convert electricity based on wind power and solar power to carbon based energy carriers could offer an interesting option. Such technologies could serve different purposes in the future energy and transport system:

- Produce a carbon based energy carrier (methane) without using fossil fuels or biomass.
- Use electricity in times with abundant wind or solar production and “store” this electricity indirectly in the form of gas.

- Act as a flexible consumer and thereby helping the wind-dominated electricity system in keeping the balance between supply and demand.
- Supply a non-fossil transport fuel

Biogas

Biogas is a gas where methane (55% - 70%) and CO₂ (45% - 30%) are the main components. Biogas is the result of microbiological processes under anaerobic conditions. Typical substrates are fats, proteins and carbohydrates.

In Denmark today there are more than 100 full-scale biogas plants running, mainly on manure, sewage sludge and biological waste from industries.

The Danish Energy Agency has estimated a total potential of biogas production in Denmark as high as app. 40 PJ based on waste products from agriculture, industry and households. In a study for Energinet.dk Ea Energy Analyses has evaluated the costs of producing biogas in comparison with the socio economic value. The total cost including upgrading to the natural gas network is approximately 140 DKK/GJ, if most of the gas is based on waste products from agriculture. This figure just about equals the socio-economic value. This value comprises benefits in the agriculture and industry and the pure energy and CO₂-value of the gas in comparison with natural gas.

3.2 Objective and goal

The objective of this WP is to analyse the “system value” of a SOEC (and methanator) plant transforming the CO₂ content in biogas to methane. In the analysed case the SOEC plant is integrated in a biogas system where the biogas is produced, upgraded and injected into the existing natural gas network.

The objective is to investigate how main parameters influence the socio-economic viability of the SOEC plant in relevant biogas-configurations.

Goal

More specifically, the purpose of this work package is to analyse:

- What process structure is the most cost-effective for an SOEC system?
 - Introducing the biogas before or after the SOEC unit.
- What could be the socio economic benefits?
- Under which conditions will the technology be commercially attractive in 2020 and 2035?

3.3 Scope

The scope of this work package is to analyse the integration of a biogas plant combined with a SOEC unit into the energy system and to calculate the socioeconomic consequences of doing so.

To do so relevant energy system parameters are analysed to find their impact on the socio economy value of a SOEC/biogas plant. Some of the most relevant parameters determining if it is beneficial to include SOEC in the energy system are:

- Cost of electricity and variation in prices
- Value of regulating power and ancillary services
- Value of produced methane (SNG)
- Fuel cost (Biogas) or alternative cost of upgrading biogas

To shed light on this a model optimising the operation of a SOEC/biogas plant in the energy system is developed and energy system analyses are made for 2020 and 2035.

Methodology

Excel model	Initially a simple model was made in Excel to get a better understanding of the relation between the most important parameters. The model can calculate yearly socio economic profit for a biogas-SOEC unit, based on inputs regarding value of flows in and out, specific investment costs for each component, size of each component and number of operational hours per year. In this model the relative sizes of the components was an input and the number of operational hours per year was an input. By changing input parameters in this model, a better understanding of the interactions in the system was obtained.
Identifying parameters	Input parameters, like specific investments for each major component, relative flows, value of flows in and flows out of the system etc. are collected from different sources. Relative flows and specific investments for SOEC and methanator are stated by Haldor Topsøe. Other flows are collected from different public sources.
GAMS optimisation model	Based on the knowledge gained from using the simple excel model, a more complex optimisation model was created. This model is made in the high-level modelling system GAMS, and is able to optimise the sizing of the components in the system as well as the operation of the system.

The model is used for several purposes:

- To compare socio economy for “traditional” upgrading of biogas with upgrading of biogas using a SOEC unit and hereby producing additional methane (In this report the SOEC-methane is called Substitute Natural Gas, SNG).
- To compare two different system layouts for a biogas-SOEC plant
- To determine optimal relative sizing of SOEC, hydrogen storage, biogas storage and methanator for the years 2020 and 2035
- To optimise operation profile for the years 2020 and 2035

4 Framework assumptions

The economic value of establishing a SOEC plant for upgrading biogas is dependent on the development of electricity and fuel prices. How these prices will develop in the future depend on a number of factors such as the development of new technologies, access and availability of energy resources at the global and regional level, economic growth and not least the policies taken to deal with climate change and to improve security of energy supply.

4.1 Climate and energy policies

While global leaders are still struggling to reach binding agreements to reduce greenhouse gas emissions ambitious targets have been formed at the EU level as well as in Denmark.

EU energy policy
measures and targets

In October 2009 the European Council agreed to set out a long-term objective to reduce the emissions of GHG by 80-95 % in 2050 compared to 1990 levels. In March 2011, this decision was followed by “A Roadmap for moving to a competitive low carbon economy in 2050”, showing possible actions up to 2050 which could enable the EU to reduce greenhouse gas emissions in line with the 80 to 95 % target. The road-map shows that electricity is likely to play a central role in the low carbon economy. By 2050 CO₂-emissions can be almost totally eliminated offering the prospect of only partially replacing fossil fuels in other sectors, such as the transport sector where the alternatives are less obvious.

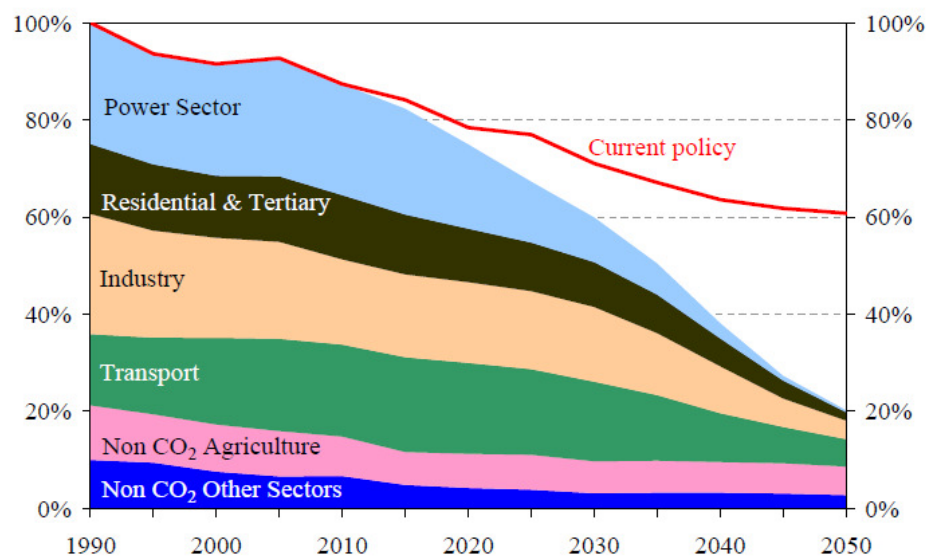


Figure 3: A pathway for reducing greenhouse gas emissions in the EU (“A Roadmap for moving to a competitive low carbon economy in 2050”, COM(2011) 112 final)

In December 2011 the Climate Road Map was followed by the Energy Roadmap 2050. By combining in different ways four main decarbonisation routes (energy efficiency, renewables, nuclear and CCS) the energy road-map explores how Europe's energy production could become almost carbon neutral.

2020 targets

In the short-term perspective to 2020, the EU target is to improve the energy efficiency by 20 %, reduce greenhouse gases by at least 20 % and increasing the share of renewable energies in the energy consumption by 20 %. In connection with COP 15 the EU made a conditional offer to the Copenhagen Accord to increase the reduction target for 2020 to 30 % depending on the international negotiations.

The 2020 targets have been transformed into concrete policies and regulation committing the EU countries to act. The emission trading scheme (EU ETS), which covers the majority of the fossil fuel power plants in the EU as well as the energy intensive industry, is one of the important tools. By 2020 all companies encompassed by the EU ETS should on average reduce their emissions by 21 % compared to 2005. Another important tool is the national renewable energy action plans requiring all member countries to set targets and implement policies to increase their share of renewable energy.

Danish policies

In Denmark the government has a vision of an energy and transport sector using only renewable energy by 2050. The Danish Climate Commission was established by the previous government and released their final report in September 2010. In this report the Danish energy system was modelled in detail, and different fossil free scenarios were presented. In all these scenarios especially wind power, but also biomass are important contributors to the fossil free energy system in 2050.

In Marts 2012 a new energy agreement was reached between a vast majority of parties in the Danish Parliament setting the scene for the next 8 years. Among other things the agreement includes a target to increase the share of wind power to 50 % of electricity demand by 2020 and to change from coal to biomass at the large combined heat and power plants.

Also, the recent energy agreement improves the framework conditions for biogas production with the aim of increasing the use of biogas. Particularly, the incentives to upgrade biogas for injection in the natural gas grid have

been improved. The specific biogas incentives provided in the agreement are described in Appendix A.

4.2 Development in fuel and CO₂ quota prices

The fuel prices of oil and gas in this study are based on the IEA New Policies Scenario as presented in IEA World Energy Outlook 2011. The New Policies Scenario, dealing with the period 2011- 2035, assumes that current G20 low carbon agreements are implemented.

The global efforts to combat climate change will reduce the demand for fossil fuels at the global level compared to a development with no low carbon regulations. Therefore, according to the International Energy Agency (IEA), increases in prices of coal, oil and natural gas will be relatively moderate. In 2035 the price of crude oil is projected to reach \$120 per barrel (in year-2010 dollars).

The price of natural gas has been decreased in the World Energy Outlook 2011 by approximately 10 % compared to the World Energy Outlook 2010. This is mainly due to the raise in expectations to unconventional gas, such as shale gas.

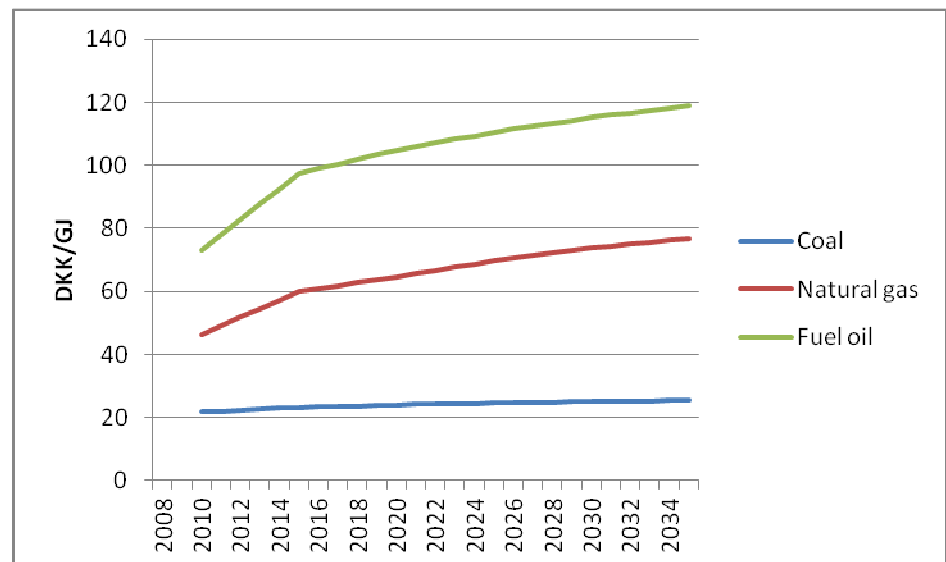


Figure 4 Fossil-fuel price assumptions in the World Energy Outlook New Policies Scenario (IEA, 2011).

WEO11 also forecasts the CO₂ price in the EU ETS. This is, in line with the above fuel prices, based on the New Policies scenario.

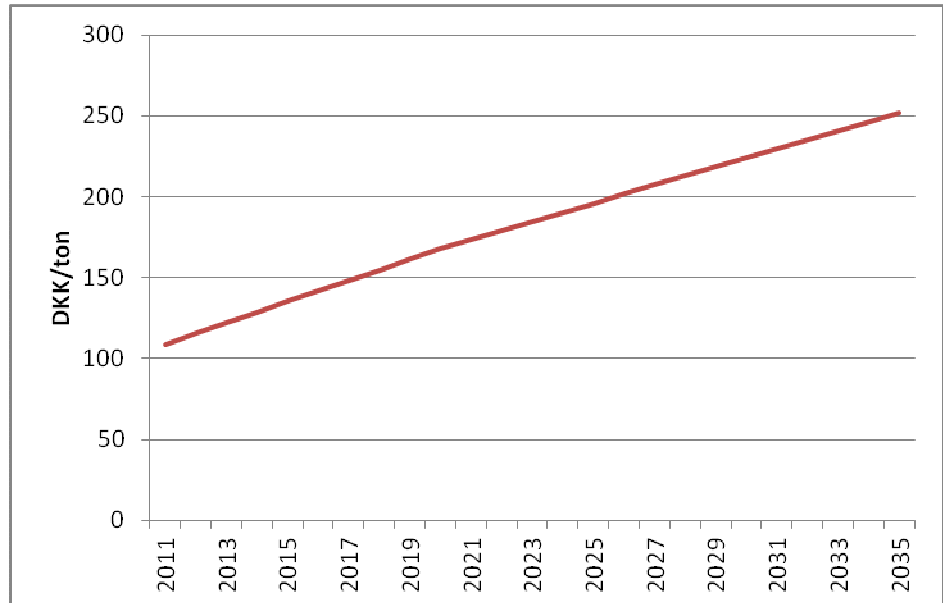


Figure 5: CO₂ price (DKK/ton) assumptions in the World Energy Outlook New Policies Scenario (IEA, 2011).

4.3 Development in the electricity market

The Nordic electricity market has developed step by step since the common exchange Nord Pool was established for Norway and Sweden in 1996. The development and integration of the Nordic electricity markets has resulted in the removal of barriers to cross-border trade and to a certain extent harmonization of rules and regulations.

Liberalised electricity markets can send strong price signals in times of scarcity and abundance. This leads to periods with price peaks and, and in situations with abundant supply, very low prices.

Several reports have pointed to the need of increased consumer response to prices, in order to further enhance the functioning of the electricity market. Consumer response means, that electricity consumers increase their demand when prices are low, and decrease demand when prices are high.

In this study the functioning of the international electricity market is basically assumed to be like it is today. The electricity price in each hour will be determined by the marginal cost of the most costly production unit running, taking congestion in the transmission system into account.

Danish Climate
Commission

The structure of consumption and production is based on scenarios developed for the Danish Climate Commission for 2020 and 2050. These scenarios were carried out with the Balmorel model, which includes the combined electricity and heat system of the Nordic countries and Germany.

The 2035 prices have been produced by taking the average of the ordered power-spot prices from 2020 and 2050 of the Nordic region. The power-spot prices reflect western Denmark.

The tariff for distributing electricity (120 DKK/MWh) is not included in the socio economic analysis.

2020 electricity system

In 2020 wind power generation equals 50 % of Danish electricity demand. This corresponds to an increase in wind power generation from approx. 7 TWh in 2009 to 17 TWh in 2020 primarily through offshore wind park development. In addition the majority of the Danish coal fired power plants will be rebuild to biomass and biogas will play an important role in the decentralized CHP systems. The neighbouring countries are expected to fulfil their commitments in the national renewable energy action plans. This development will result in increased fluctuations in electricity prices. Some measures to decrease these fluctuations are also implemented in the simulation, including an increase in transmission capacity to the surrounding countries and an increased consumption of electricity for the production of district heating.

2050 electricity system

The 2050 system is also based on a high wind power penetration in Denmark. More than 75 % of the Danish electricity generation will be based on wind power. The electricity generation of the surrounding countries (Germany, Norway, Sweden and Finland) is depicted in the figure below.

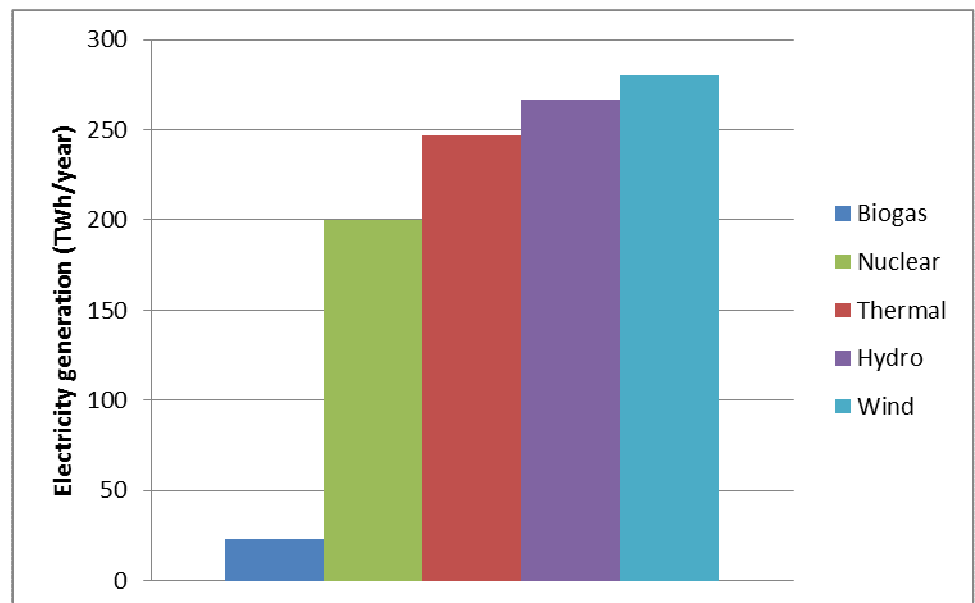


Figure 6: Electricity generation in 2050 in Germany, Sweden, Norway and Finland (TWh/year)

Table 3 shows the capacities of the power plants in the Danish electricity and heat system in 2050 distributed on fuels.

(MW)	Installed capacity
Waste	603
Straw	17
Wood chips	100
Biogas	309
Biogas (upgraded) (peak power and back-up)	5.121
Wind	21.559
Solar	5.500
Wave power	1.400

Table 3: Capacity in MW in the Danish electricity and heat system in 2050

4.4 Development of biogas in Denmark

The biogas process has been used for more than 100 years in the waste water treatment plants. There are more than 65 plants in Denmark with decay tanks and biogas production. A few industries and waste disposal sites also utilises the biogas¹. These sources are considered fully utilised and the potential is therefore within the agricultural sector and biogas plants based on slurry.

In total the domestic animals within the agricultural sector produce 30 million tons of slurry a year. Only 5-7 % of this is utilised for biogas production. Today the biogas is primarily used at local CHP plants.

In 2010 the Danish Energy Agency estimated the total biogas production as shown in the table below. The table also illustrates another estimate of the potential carried out by PlanEnergi for the Danish TSO Energinet.dk in the report “Biogaspotentiale i danske kommuner”.

¹ Danish Energy Agency: Anvendelse af biogasressourcerne og gasstrategi herfor, notat, maj 2010

PJ	Potential	Potential	Production 2008
	PlanEnergi	DEA	DEA
Animal manure	23	26,0	1,06
Waste water sludge	2-4	4,0	0,84
Industrial waste, Danish	2	2,5	1,04
Industrial waste, import			0,65
Meat and bone products	0,53	2,0	0,03
Municipal waste		2,5	0,04
Garden and park waste	1	1,0	0
Landfill gas		1,0	0,27
Energy crops	42		
Meadow grass	3		
Catch crops	14		

Table 4: Potential and estimated biogas production, DEA, 2010 and PlanEnergi, 2010.

The current production of approx. 4 PJ/year corresponds to approx. 0.5 % of the Danish gross energy consumption. The total potential of 40 PJ/year can therefore cover approx. 5 % of the current gross energy consumption.

The graph below shows the development in the Danish biogas production between 2000 and 2009. It appears that the biogas production has increased steadily, but at a moderate pace.

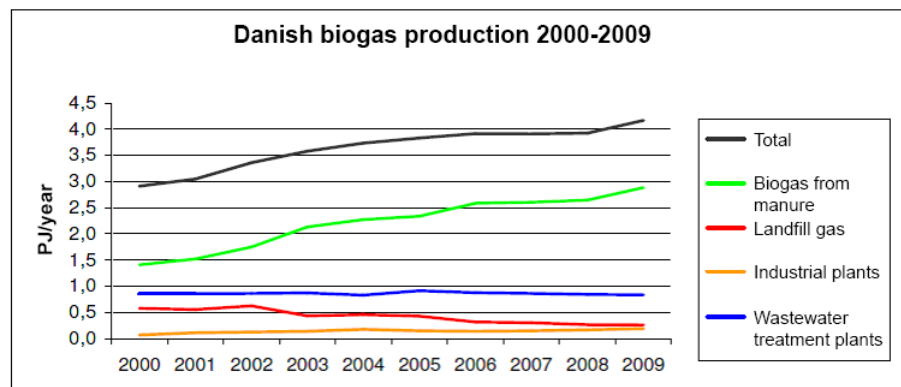


Figure 7: Development in the Danish biogas production in the period 2000-2009. DEA, 2010.

5 Two SOEC configurations

A system consisting of a biogas unit and a SOEC unit can be combined in different ways. In this project two configurations are tested;

1. A configuration where biogas from the biogas plant is mixed with hydrogen from the SOEC in a methanator producing SNG
2. Another configuration where biogas from the biogas unit is introduced directly into the SOEC unit (after purification of the biogas)

In this report we analyse, which of the two process structures is the most cost-effective, when looking at the plant integrated in the energy system.

Regulating abilities

TOFC (Topsoe Fuel Cells) has recently tested the dynamic abilities of the SOEC in a project together with RISØ and Aalborg University (Energinet 2011-1-10609). The conclusion was that apparently the SOEC stack sees no problem in fast change of current, when the temperature is kept stable.

5.1 Description of components

SOEC unit

In the solid oxide fuel cell steam (water vapour) can be split into hydrogen and oxygen or carbon dioxide can be split into carbon monoxide and oxygen. Both processes consume power. One of the advantages of the SOEC is that it is almost thermo neutral, meaning that there is little excess heat/loss from the process, and therefore most of the energy in the power used for splitting is converted to chemical energy in the product gas. This results in a high efficiency.

Other advantages of the SOEC are:

- the possibility of co-electrolysis of steam and carbon dioxide
- it is expected to have high efficiency also at part load
- it is expected to have good dynamic properties, and could potentially be run in reverse mode with production of electricity

Configuration 1

In configuration 1 only steam will be introduced in the SOEC and split into hydrogen and oxygen.

Configuration 2

In configuration 2 biogas (mainly consisting of methane and carbon dioxide) and steam is introduced in the SOEC. The methane will pass unchanged, and

the steam and the carbon dioxide will be split into hydrogen, carbon monoxide and oxygen.

The oxygen is taken out of the process, and not used for any purpose in these cases, but the other gas-components are sent to storage or directly to the methanator.

Methanator

In the methanator a catalytic process shifts the gasses to produce as much methane as possible limited by equilibrium constraints. The process differs from configuration 1 to 2, since the gas composition is not the same in the two configurations, when entering the methanator. The process is exothermic, and some of the produced heat can be used either to produce steam for the SOEC, or excess heat can be used for district heating or as process heat in the biogas plant if the biogas plant is located in the vicinity of the methanator unit.

The dynamic properties of the methanator are expected to be good.

5.2 Configuration 1

In configuration 1 biogas from the biogas plant is mixed with hydrogen from the SOEC in a methanator producing SNG:

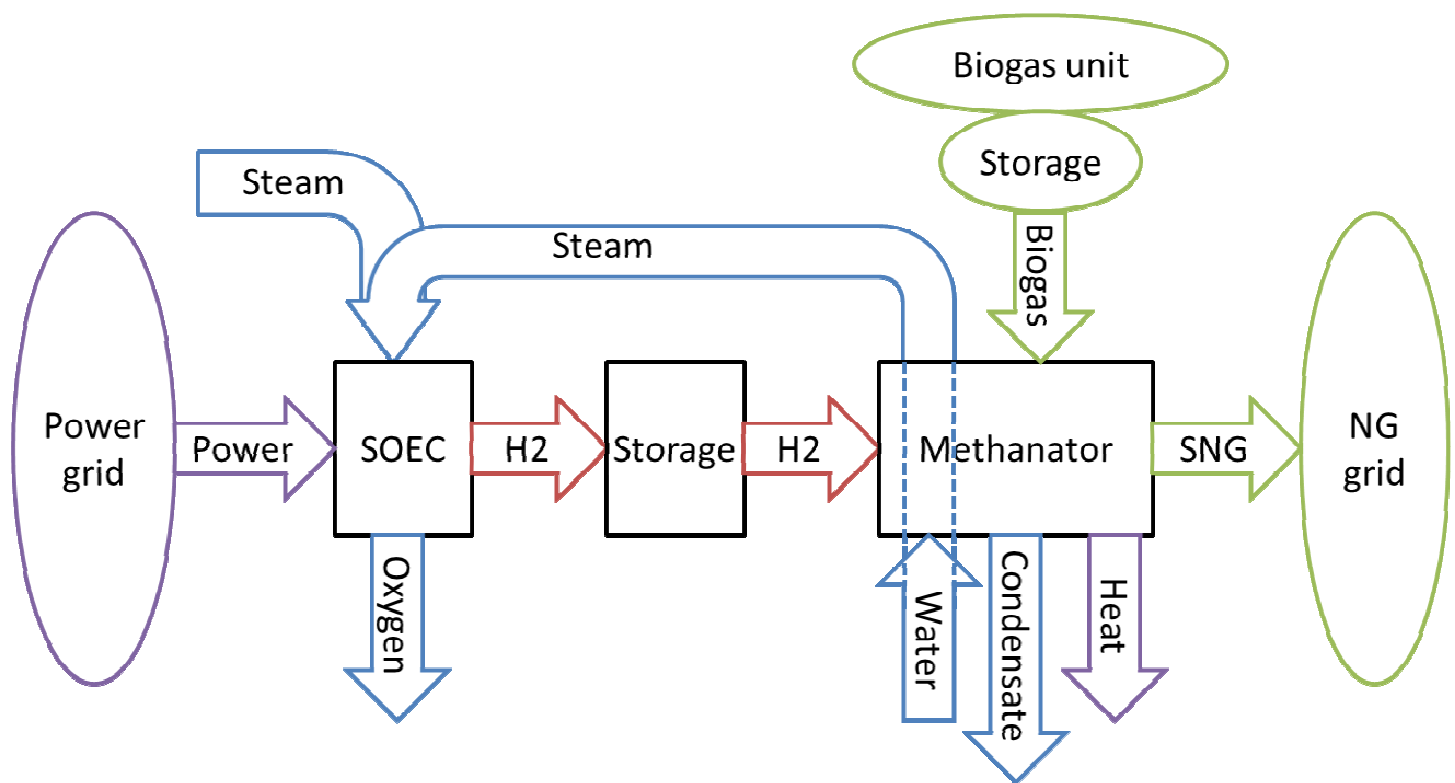


Figure 8: Concept design for SOEC plant integrated in the energy system. System template for process where biogas is mixed with hydrogen in the methanator.

Need for storages

For a system where biogas is mixed with hydrogen in the methanator, we do not see the need for a biogas storage, since the methanator will be in operation in the same hours when the biogas unit is. Since the biogas unit will be in operation all year round, so will the methanator. In order to utilise the hours with cheapest electricity for the high electricity demand in the SOEC, the SOEC unit is not expected to operate all hours of the year, and thus there is a need for a hydrogen storage.

The same might not be the case for the other system configuration where biogas is introduced in the SOEC unit.

5.3 Configuration 2

In configuration 2 biogas from the biogas unit is introduced directly into the SOEC unit after purification of the biogas:

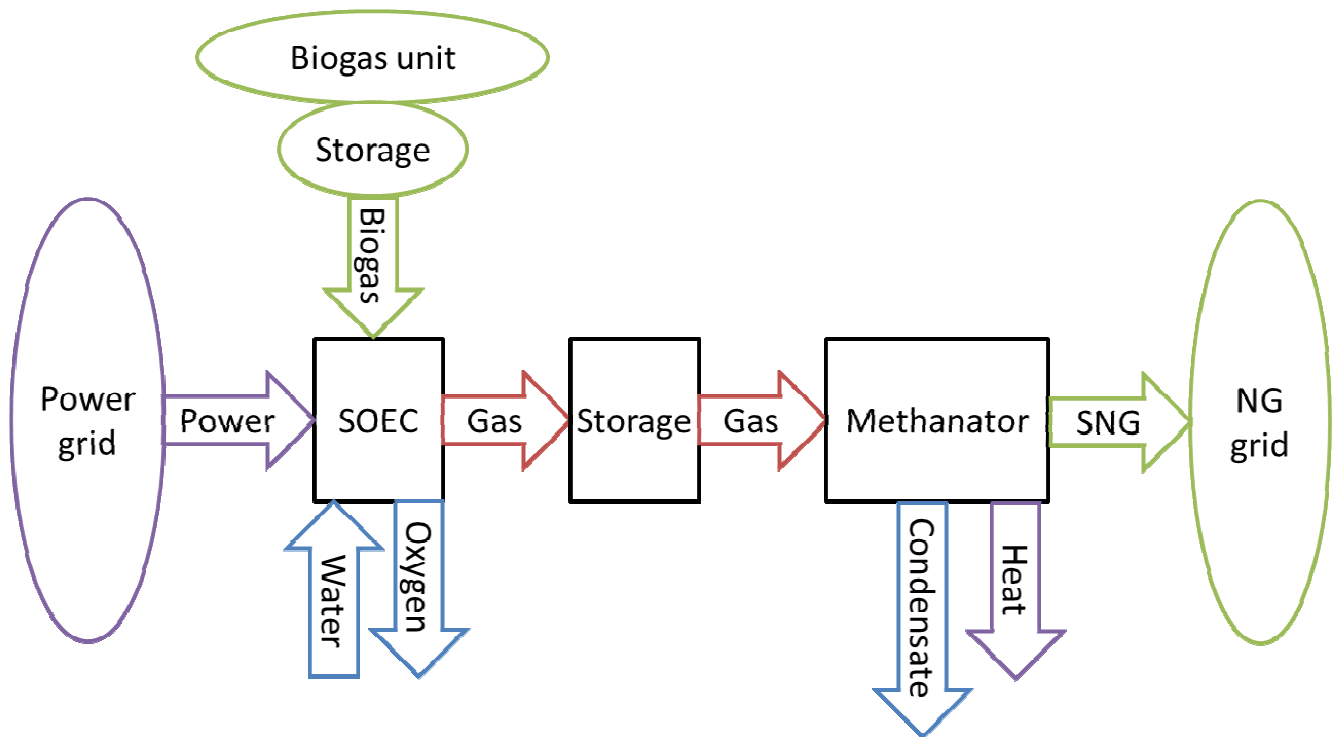


Figure 9: Concept design for SOEC plant integrated in the energy system. System template for process where biogas is introduced in the SOEC unit before the methanator.

Need for storages

A biogas storage is expected to be advantageous in this configuration because – due to the fluctuations in the electricity prices – it is not desirable that the SOEC unit is in operation all year round. For a system where biogas is introduced in the SOEC, the hydrogen/syngas storage is not required to make the SOEC change load, but the hydrogen/syngas storage might still be relevant in order to invest in a smaller methanator and prevent the methanator from changing load all the time.

5.4 Identification of critical parameters

When analysing the economics for a SOEC unit in combination with a biogas plant, the most important parameters are the investment costs for the different main components, the ratio between different flows in/out of the system (efficiency) and value of different flows in/out of the system.

Most relevant parameters

The parameters which are presumed to be most relevant for the economic calculations are:

Investments

- SOEC unit
- Methanator

- Hydrogen storage
- Biogas storage

Flows

- Biogas
- Hydrogen from SOEC
- SNG from methanator
- Power used in SOEC
- Water/steam for electrolysis
- Heat production (from SOEC and methanator)
- (Oxygen from SOEC)
- (Condensate from methanator)

Value of flows

- Biogas
- SNG
- Power
- Heat production

6 Description of cases

A case with a Biogas/SOEC plant producing/using 7.500.000 Nm³ biogas/year or 860 Nm³ biogas/h is analysed in this project. In the future bigger biogas plants might be a reality, and some benefit of scale is therefore studied in the sensitivity calculations later.

The number of operational hours for the biogas plant is expected to be 8.500 h/year.

6.1 Flows

The relative flows in and out of each component differ between configuration1 and 2.

Configuration 1

The flows for a plant where SOEC and methanator are dimensioned for 8.500 h/year in combination with the above described biogas plant, will in configuration 1 be:

		Mass Flow Kg/h	Volume Flow Nm ³ /h	Energy Flow MWh/h
IN		969	1.206	4,54
Steam	H ₂ O	969	1.206	0,63
Power				3,91
OUT		-969	-1.808	-4,54
Hydrogen	H ₂	-109	-1.206	-3,61
Oxygen	O ₂	-860	-603	
Heat (DH+loss)				-0,93
		<i>DH</i>		-0,60
		<i>Loss</i>		-0,32

Table 5: Flows (mass, volume and energy) in and out of SOEC.

When optimising the relative sizing, some components might be bigger and thus run less than 8.500 h/year. In that case the flow figures in Table 5 and Table 6 will be scaled to the optimal absolute size.

Electricity consumption
in SOEC

The electricity consumption in the SOEC is according to Haldor Topsøe 3.15 kWh/Nm³ H₂. In addition to this, 2-3 % extra electricity is used for preheating, compression of biogas and compression of SNG. We assume that

the SNG will be delivered to the natural gas grid at 40 bar, and the electricity consumption for compression to 40 bar is thus included.

Heat (District heat/loss) from SOEC

It is also assumed that 65 % of the heat from the process in the SOEC can be utilised, e.g. for district heating. The remaining 35 % are losses.

Mass flows in SOEC

The hydrogen production in the SOEC is determined from the hydrogen need in the methanator to convert all CO₂ in the biogas to SNG. The other flows are calculated from the hydrogen flow.

SOEC load changes

The SOEC can perform quick load changes from 0 to 100 % if it is kept warm. In the modelling we anticipate that the SOEC is always kept warm, but the (small) energy consumption to do this, is not included in the model. Therefore the SOEC can change load in the model from 0 to 100 % without any change in efficiency.

		Mass Flow Kg/h	Volume Flow Nm ³ /h	Volume Part %	Energy Flow MWh/h
IN		2.069			9,15
Biogas		991	859	100	5,54
	CO ₂	587	299	35	
	CO	5	4	1	0,02
	H ₂	-	-	0	-
	CH ₄	398	556	65	5,53
	H ₂ O	-	-	0	-
Hydrogen	H ₂	109	1.206		3,61
Water		969			
OUT		-2.069			-9,15
SNG		-625	-884	100	-8,52
	CO ₂	-12	-6	1	
	CO	-	-	0	-
	H ₂	-2	-25	3	-0,08
	CH ₄	-607	-848	96	-8,44
	H ₂ O	-3	-4	0	
Condensate	H ₂ O	-475	-591		
Heat (steam+DH+loss)		-969	-		-0,63
	Steam	-969			-0,63
	DH				-0
	Loss				-0

Table 6: Flows (mass, volume and energy) in and out of methanator.

Mass flows in methanator

The ratio between biogas, hydrogen, SNG and condensate is stated by Haldor Topsøe. The gas composition in the biogas and in the produced SNG is stated by Haldor Topsøe.

Heat (steam) from methanator

The heat produced in the methanator is used to evaporate water and heat up the steam used in the SOEC. We assume that the steam production from the methanator is just exactly enough to support the SOEC with sufficient steam. This means that no excess heat is produced in the methanator besides the steam for the SOEC.

The minimum load of the methanator is 20 %.

The process and efficiencies and hereby the flows are not expected to change significantly from 2020 to 2035.

Configuration 2

The flows in configuration 2 for a plant where SOEC and methanator are dimensioned for 8.500 h/year in combination with a biogas plant, are displayed in Table 7:

	Mass Flow Kg/h	Volume Flow Nm3/h	Energy Flow MWh/h
IN	1.823	1.895	9,55
Biogas	988	857	5,53
Steam	835	1.038	
Power			4,02
OUT	-1.820	-1.901	-9,55
Gas	-629	-885	-8,56
Oxygen	-857	-600	
Condensate	-335	-416	
Heat (DH+loss)			-0,99
<i>DH</i>			-0,63
<i>Loss</i>			-0,36

Table 7: Flows (mass, volume and energy) into SOEC and out of SOEC/methanator in configuration 2.

When optimising the relative sizing some components might be bigger and thus run less than 8.500 h/year. In that case the flow figures in Table 7 will be scaled to the absolute size.

Heat (District heat/loss). Load changes for SOEC As in configuration 1 we assume that 65 % of the heat from the process in the SOEC/methanator can be utilised, e.g. for district heating and we also assume that the SOEC can change load in the model from 0 to 100 % without any change in efficiency.

In the methanator the gas composition is changed to SNG containing close to 100 % methane, to be fed into the natural gas grid. Subsequent addition of propane is not considered.

The minimum load of the methanator is 20 %.

The process and efficiencies and hereby the flows are not expected to change significantly from 2020 to 2035.

6.2 Investments

Investment in SOEC Haldor Topsøe has estimated the investment in the SOEC and the auxiliary equipment to be 1000 Euro/Nm³ hydrogen in 2020 for a plant of this size (7.500.000 Nm³ biogas/year), when biogas is not introduced in the SOEC. If introducing biogas in the SOEC, the investment is estimated to be 25 % higher because there is a need for more cells. This is mainly due to expected higher degradation rate from sulphur in the biogas and larger heat exchangers.

Investment in methanator Haldor Topsøe estimated that the investment cost for the methanator and its auxiliary equipment is approx. 20 MDKK in 2020 for a plant of this size (23.000 DKK/Nm³ SNG produced), when biogas is not introduced in the SOEC, but mixed with hydrogen in the methanator. If introducing biogas in the SOEC, and thereby receiving a mixed gas in the methanator, the investment in the methanator is estimated to be 20 % lower because the methanation reactor becomes smaller due to better reaction kinetics.

Investment in hydrogen storage For this project two technologies for hydrogen storage are assessed: A possible hydrogen storage in a cavern is in one reference estimated to cost 10 Euro/kWh in the size of 10 MWh and the pressure 30 bar². Auxiliary equipment is assumed not to be included in this price, and is estimated to add an additional 25 % to the investment cost resulting in a total investment cost of 93 DKK/kWh. A hydrogen storage in a big pressurised steel tank is estimated to cost 40 DKK/MJ in 2020 (or 144 DKK/kWh) for a tank of the size

² Technology data for Energy Plants, Danish Energy Agency and Energinet.dk, June 2010

50-100 GJ and the pressure 10-15 bar³. The hydrogen storage is assumed to cost 100 DKK/kWh in the modelling.

In configuration 2 the hydrogen storage is replaced by a gas storage for the mixed syngas mainly consisting of methane, hydrogen and carbon monoxide. We assume that the investment cost is the same as for a hydrogen storage, when measured on volume base.

Investment in biogas storage

A biogas storage containing around 6-8 hours of biogas production, which is around 5-7.000 Nm³ is estimated to cost 250 DKK/Nm³. A biogas storage is not expected to be build larger than for 12 hours load (10.000 Nm³).

The investment costs are not expected to change significantly from 2020 to 2035.

6.3 Other financial input data

We apply a 5 % annual interest rate in real terms.

The lifetime of the equipment is expected to be 5 years for SOEC (due to degradation of cells) and 20 years for all other equipment.

The annual operation and maintenance costs are assumed to make up 5 % of the investment.

6.4 Value of different flows

The economic values of the different flows used in the model are listed below. The assumptions underlying these values for biogas, upgrading of biogas, natural gas and CO₂ quotas are described in chapter 4.

³ Scenarier for samlet udnyttelse af brint som energibærer i Danmarks fremtidige energisystem, RUC, 2001, Bilag A: Teknologikatalog

		2020	2035
Biogas	DKK/GJ	115	132
Upgrading of biogas	DKK/GJ	25	25
Heat	DKK/GJ	75	75
Natural gas	DKK/GJ	64,7	76,8
CO2 price	DKK/ton	168	252
CO2 price	DKK/GJ	9,5	14,3
Bio-SNG	DKK/GJ	140	157
CO2-SNG, low	DKK/GJ	74,2	91,2
CO2-SNG, high	DKK/GJ	140	157

Table 8: Value of different energy flows in/out of the modelled unit used for modelling the economy in 2020 and 2035.

Electricity

For the modelling a forecast of the power spot prices for each hour for 2020 and 2035 are needed. These forecasts are based on scenario calculations made in Balmorel for 2020 and 2050 to The Danish Commission on Climate Change Policy. The analyses were made in 2010 by Ea Energy Analyses.

To make a forecast for 2035 electricity prices, an average of the sorted prices for 2020 and 2050 are used.

Time series for 2020 and 2050 from the Danish Commission on Climate Change Policy

The electricity prices are calculated in Balmorel as market prices on the wholesale market. These prices can be interpreted as the societal value of electricity at a given time. The prices ignore certain grid tariffs (operations and investments in the distribution grid), distribution losses and "PSO" tariffs to support renewable electricity. There is no certain blueprint for how to recover grid costs and PSO tariffs in a dynamic electricity system with high wind penetration. It is possible, that flexible electricity consumers in reality will not incur extra costs in the system. Therefore the calculated electricity price in Balmorel is probably a fair proxy for the total socio economic electricity price for these kinds of consumers. Cost recovery of PSO and distribution grid is not further analysed in this project.

Construction of time series for 2035 power prices

The 2035 electricity prices follow the yearly profile of the 2050 electricity prices of the western region of Denmark, but are damped using the average sorted fluctuations of the 2020 and 2050 electricity prices of the same region.

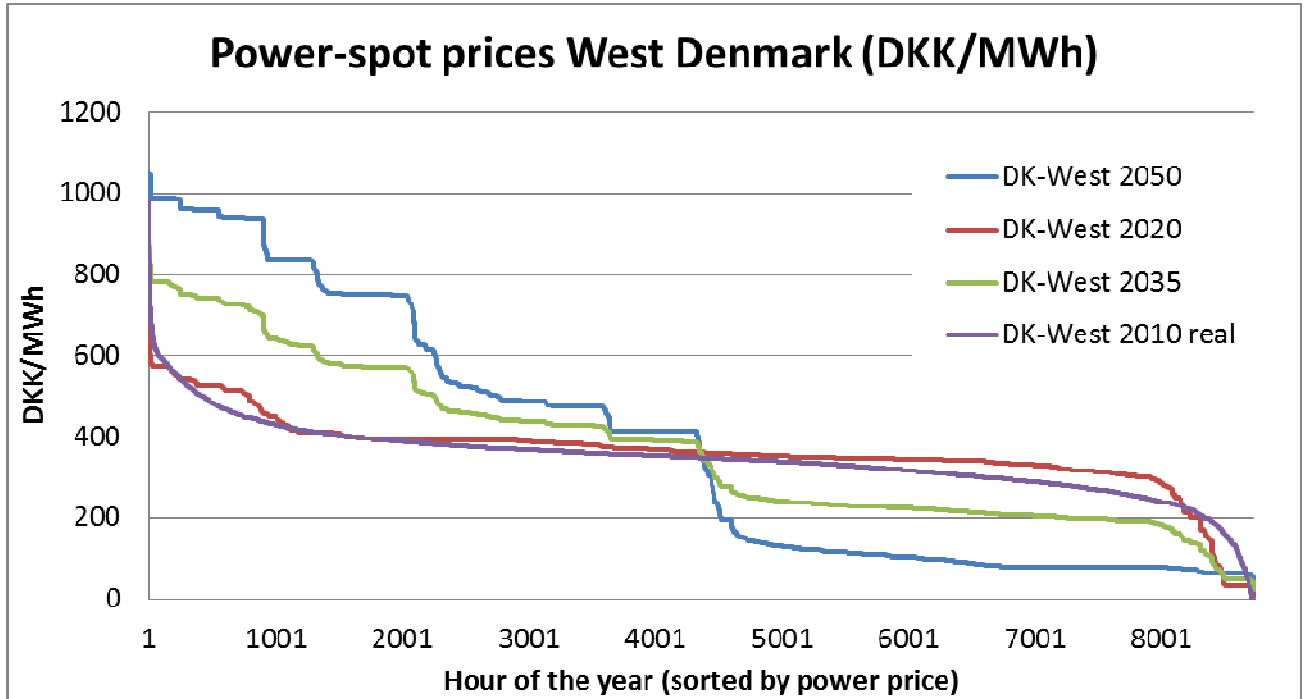


Figure 10: Power prices for 2010 (real spot price), 2020, 2050 (from the Climate commission modelling) and 2035 (calculated as average of sorted prices for 2020 and 2050). Prices are for Western Denmark and are in DKK/MWh.

Heat

The excess heat from the methanator/SOEC can be utilised for district heating or for process heat in the biogas plant.

Value of heat

The value of heat is in this project determined by the alternative heat production cost. In this case the long term marginal cost of a wood chip fired boiler is chosen as the alternative. Based on calculations in the ENERCOAST⁴ project, the value of heat is estimated to be 75 DKK/GJ. This value is also used for 2035.

Methane (SNG)

Methane is the dominant component in natural gas. The value for society of producing methane from CO₂, could be calculated as the replacement value of natural gas comprising both the energy value and the CO₂ value. In other

⁴ Biomassehandlingsplaner for Randers, Norddjurs og Syddjurs, Enercoast slutkonference 27. juni 2012 Jesper Werling, Ea Energianalyse

words, the production of methane simply saves the cost of natural gas and of buying CO₂ quotas.

However, in a more long-term and broad evaluation there are some challenges to such a simple perspective:

- Society aims at reducing the use of fossil fuels to zero. If this goal is pursued in a larger scale, the price of natural gas must be expected to drop to a very low level (supply/demand balance). Due to this feedback mechanism it will not be relevant to use the price of natural gas as a basis for the evaluation of the value of sustainable methane in a “non fossile” future.
- In the longer term the SOEC-methane will in practice displace biomass-based gases, e.g. biogas and gasification gas. In a future where biomass is a scarce resource, it is the cost of these gases that represent the real saved costs to society.
- It can be disputed whether the projected CO₂ price in the EU-ETS represents the real abatement cost of CO₂ in an ambitious reduction scenario.

When society moves towards a fossil free future, it seems wrong to continue to use the price of fossil fuels as the main value yardstick for the replacement fuels. When the overall political goal is decided (zero fossil fuels in 2050), the important question is what the most economic path will be. To answer this question it is necessary to compare the costs of different “sustainable fuels”.

Such a comparison should include a range of relevant alternative fuels each described by their main cost elements including: production costs (opex & capex), emission costs/benefits and resource costs/benefits. It has not been possible within the scope of this project to undertake such calculations with any reasonable precision. A more simple methodology has been chosen, based on previous calculations of the socio economic value of biogas.

Value of biogas

The value for society of producing biogas is shown in “Biogas - analyse og overblik, Ea Energianalyse, 2012”. In addition to the basic replacement-value of natural gas and saved CO₂ emission, a range of benefits for the farmer and industry are included. These benefits are mainly linked to the handling of manure.

We have on this basis calculated two values for SOEC methane: An upper value based on the previous calculations of the value of biogas and a lower

value based only on the price of natural gas and of CO₂. Below these two values are shown for the year 2020.

High/low value of SOEC-methane (CO₂-SNG)

- a) High value: The value equals the societal value of upgraded biogas (115 DKK/GJ + 25 DKK/GJ = 140 DKK/GJ in 2020)
- b) Low value: The value equals the value of natural gas + avoided CO₂ (64,7 DKK/GJ + 9,5 DKK/GJ = 74,2 DKK/GJ in 2020)

In 2035 the low value increases to 91 DKK/GJ as both the price of natural gas and the price of CO₂-quotas are expected to increase. The high value is projected to remain unchanged between 2020 and 2035.

Value of upgraded biogas (Bio-SNG)

The SNG from the methane in the biogas (here called bio-SNG) has the same value as upgraded biogas.

Additional steam

The SOEC unit needs steam as an input to produce hydrogen and water. When the SOEC and the methanator are of the same size, the methanator produces enough heat to evaporate enough water and hereby produce enough steam for the SOEC. The SOEC and the methanator are of the same size when the SOEC produces exactly the hydrogen needed in the methanator to convert all CO₂ from the biogas to CH₄ in each hour.

Cost of producing additional steam

If the SOEC is bigger than the methanator, the methanator does not produce enough steam for free. This means that additional steam must be produced/bought. This steam could be produced by means of an electrical boiler, which has low investment costs, but on average high operation costs. Applying a biomass boiler would mean higher investment cost but lower operation costs. For the purpose of the calculations the steam price (for additional steam) is for each hour set equal to the power price for the given hour. The investment costs of the electrical boiler are considered to be negligible.

Other flows

Other flows in the model are oxygen flow from the SOEC unit, water flow to the methanator and condensate flow from the methanator. These flows are not considered significant for the economy of the plant, and the value is therefore set to 0 DKK. The condensate out of the methanator might be used as boiler feed water for the methanator steam system. This will halve the need for water and eliminate the disposal challenge. The oxygen could in some process configurations be used, e.g. in combination with biomass gasification or in CO₂- capture processes using oxy-fuel combustion. In this

case it is not obvious how to utilize the oxygen, and therefore as a conservative estimate the value is set to zero.

- Oxygen: 0 DKK/Nm³
- Water: 0 DKK/ton
- Condensate: 0 DKK/ton

7 Modelling of a biogas-SOEC unit

In order to model the Biogas-SOEC system, first a simple excel model was constructed to gain knowledge about interaction between the most important parameters. Afterwards experiences and knowledge from the excel model was used to construct an optimisation model in GAMS.

The first simple model was made in excel to gain knowledge of the relation between the most important parameters.

Input to this model is:

- the sizes of each of the components
- the number of operational hours per year
- relation between flow (energy/mass) for each component
- value of flows in and out
- specific investment costs for each component

The output from the model is the yearly socio economic profit for a biogas-SOEC unit. By changing input parameters in this model, a better understanding of the interactions in the system was obtained.

7.1 Optimisation model

A more complex optimisation model was created in GAMS. This model can optimise the sizing of the components in the system and can optimise the operation of the system including filling/emptying of storages. This model has been used to produce the final results of the work package accounted for in the report.

These following sections describe the mathematical modelling of the Biogas-SOEC system using GAMS (General Algebraic Modelling System).

Purpose of the optimisation model

The purpose of the GAMS model is to determine the optimal size of relevant system components as well as the optimal hourly operation for a given year (2020 or 2035).

The model is used for several purposes:

- To compare socio economy for “traditional” upgrading of biogas with upgrading of biogas using a SOEC unit and hereby producing additional SNG

- To compare two different system layouts for a biogas-SOEC plant
- To determine optimal relative sizing of SOEC, hydrogen storage, biogas storage and methanator for the years 2020 and 2035
- To optimise operation profile for the years 2020 and 2035

Elements in the optimisation model

The system consists of the following system component:

- Process Components: SOEC, Methanator, Biogas unit (simple model)
- Storage: Biogas storage, hydrogen storage
- Input/output values

The system is optimised with respect to investment and operating cost of all system components, the given price of fuels and other inputs (power, biogas, and steam), and the sales price of SNG, and heat. The model has full foresight of all prices for the full optimisation period (1 year).

The model calculates the operating cost for each hour of the year and summarising the economy for a whole year. The model optimises the relative sizing of the components (SOEC, methanator, hydrogen storage and biogas storage) and optimises the operation.

The inputs to the model are specific investment cost for all components, ratio between flows and value of each flow. These are further described in chapter 4 and 6.

The primary condition of the system is a fixed size of the biogas unit, and hereby a fixed hourly biogas flow (the biogas unit is assumed to be in operation 8500 h/year).

The output from the calculations is presented in an Excel- interface.

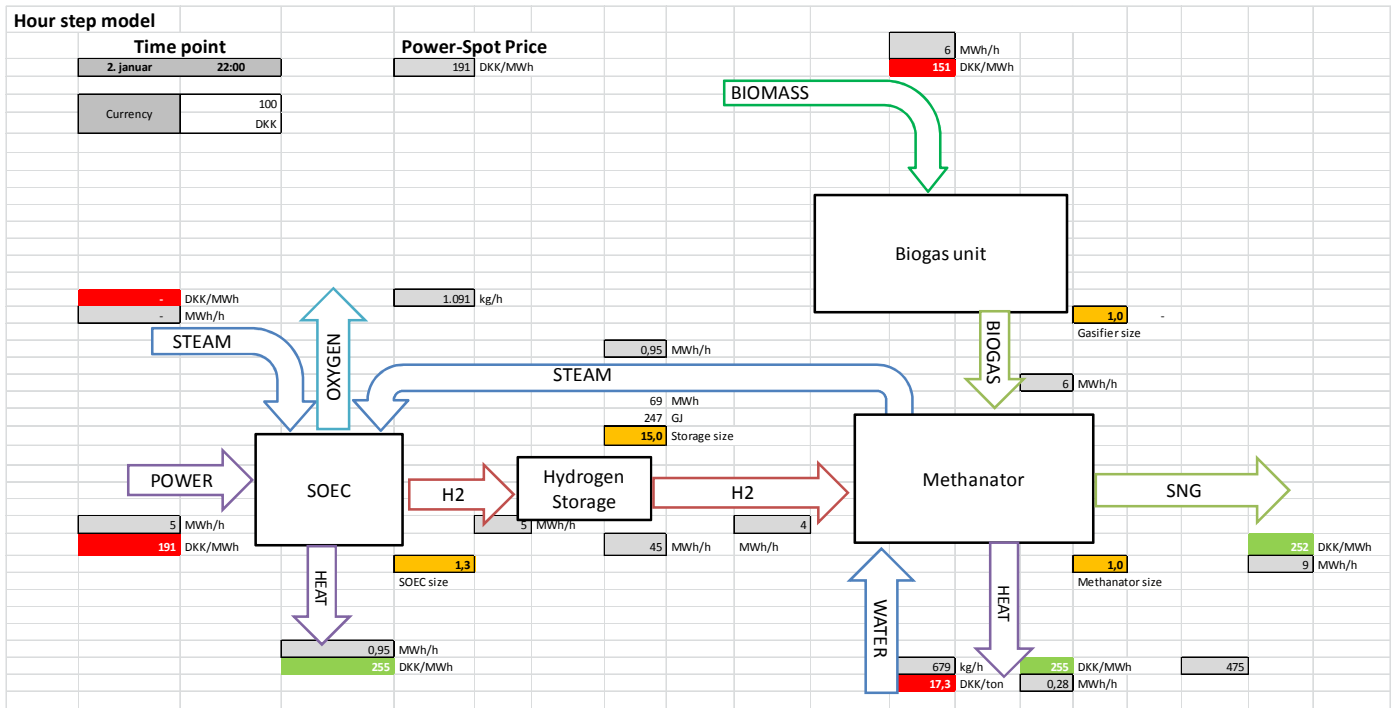


Figure 11: Interface showing output from model calculations.

The main output from the model is the yearly profit from the optimised system. Other outputs are (see also chapter 5.4):

- Sizing of each component
- Operation of storages
- Hourly and yearly value of all flows and total expenses/income

7.2 Modelling the Biogas-SOEC using GAMS

By formulating the Biogas-SOEC as a linear program (LP), different aspects of the model can be optimised with regard to maximising the economy. The LP of this report aims at optimising the operation of the plant assuming a given price of electricity and other fuels/flows and at the same time optimising the relative size of individual components of the plant given a cost of building and maintaining these components. This optimisation strategy will then give a valuable insight into how the plant components should be proportioned and what the expected output economy could be following an optimal operation schedule. The LP model is not an operation simulation but is a deterministic model with full insight into all fuel- and components costs for the total model horizon.

Building the model

In the LP model the plant components do not necessarily reflect all of the actual plants individual components. Some groups of components will be

merged into one component, simply because no extra operation information will be gained, by modelling these components apart. This simplification leaves the model having two main components; the SOEC and the methanator. Besides these main components the model can build a hydrogen-storage and a biogas-storage in order to allow non-synchronised operation of the SOEC and methanator and between the biogas unit and the methanator. All components are assumed to have a linear relation between cost and size. The plant is modelled using the concept of vertices and edges. The components represent vertices and the energy and mass flows between these components are then the edges.



Flow balance

A component is defined as having an amount of flows out of and an amount of flow into itself. These flows have to at all-time balance, such that if one flow drops all other flows drops proportionately. For single time point and component, this condition can be formulated in mathematical terms as thus:

$$F_{ij} \cdot \sum_{i \in I(j)} E_{ij} = E_{ij} \cdot \sum_{i \in I(j)} F_{ij}$$

$$\sum_i F_{i,in} = \sum_i F_{i,out}$$

Here F is a quantitative flow and E is a balance constant. The first equation ensures that the flows always balance relative to each other and the second ensures that the ingoing flow balance with the outgoing flow. The model used for optimisation contains both balance constants for energy and mass, since these two groups together will be able to represent all flows. This also implies that the model contains two sets of balance equations. Flows which do not end in a vertex, is modelled with unique LP constrains, meaning that constrains are formulated specific for each case. Storages are not considered a vertex in the model. This mean that flows in and out of storages are also modelled as unique constrains.

Component proportions

Each component is modelled with a variable size, and all flows are constrained by this component size. The mass and energy balance constants represent the unit size of a component. This means that if the component maximum allows flows matching the balance constants, the variable size of the component is 1. The components can also constrain the flows downwards. This is the case for the methanator, which as a minimum allows flows corresponding to 20% of full operation. The storages have similar constraints relating to storage content. Finally the components also have constraints on the hourly flow gradients, meaning that between two time-points the change in a given flow is downward and upward limited.

Economy

The object of the LP model is to maximize output economy, hence the model also contains constraints related to the cost of flows, the investment and operation and maintenance costs of components. Flows between components have a price of 0. Flows representing income (e.g. selling heat and SNG) have positive prices and flows representing expenses (e.g. fuel cost like biogas and power) have negative prices. The optimisation objective of the model thereby becomes the sum of income minus the fuel expense, minus the investment cost, minus the operation and maintenance cost. The investment cost is weighted according to the optimisation time horizon with respect to the lifespan of the component. This means that a component with 20 year lifespan and a one-year optimisation horizon have an investment cost of the yearly down payment of a 20 year loan including interest rates.

Electricity prices

The variable element of the LP model is the price of electricity. The electricity price is based on the hourly spot-price which means that the price fluctuates. It is this fact that constitutes the incentive to use a LP model to optimize operation and plant size. During periods of high prices the electricity usage should be low or zero and storages will then be the main source of ingoing flows to the methanator. How to determine the optimal size of these storages as well as the other component then become complicated. Using an average fixed price of electricity does not capture the economic benefits of exploiting periods of low and high electricity prices. Using the LP model does. This element of fluctuating electricity prices also becomes exceedingly important since these fluctuations becomes continuously more dominant in future model scenarios, making the plant continuously more profitable if the operation can be adjusted to these prices.

7.3 Mathematical model formulation

All these considerations imply then the following LP model of the SOEC-Biogas plant:

Sets:

- \mathcal{E} energy form/flow (consists of {power (used), power (produced), steam (free), steam (with cost), oxygen, hydrogen, water, heat, condensate, biogas (CH₄), SNG})
- \mathcal{T} time (consists of [0, T])
- \mathcal{d} system components (consists of {SOEC, Methanator, CH₄ storage, H₂ storage})
- \mathcal{d}_{y10} system components with a life span of 10 years (consists of {SOEC})
- \mathcal{d}_{y20} system components with a life span of 20 years (consists of {Methanator, CH₄ storage, H₂ storage})
- $\mathcal{d}_{process}$ system components which perform a chemical process (consists of {SOEC, Methanator})
- $\mathcal{d}_{storage}$ system components which are storage (consists of {CH₄ storage, H₂ storage})
- \mathcal{l} capacity index (consists of {min, max})
- \mathcal{n} operations cost group (consists of {group 1, group 2})
- io input/output indicator (consists of {in, out})
- u unit of flow (consists of {kg, mwh})
- c currency (consists of {Euro})

Parameters:

- $\pi_{e,t}$ spot prices of energy e at time t
- $p_{e,u}$ price of energy type e with respect unit u
- $eb_{d,io,e}$ energy balance constant of flow of energy type e for component d with direction io
- $mb_{d,io,e}$ mass balance constant of flow of energy type e for component d with direction io
- $notb_{d,io,e}$ set for including flows which are not among the balance constants
- fs_d system size capacity of component d
- $f_{d,l}$ maximum and minimum capacity of component d
- $g_{d,e}$ gradient for component d of energy type e

- us_d unit size for component d
- uc_d unit investment cost for component d
- $rate$ interest rate of financing
- $dvrates$ operations costs rate
- $npayments_n$ number of payments with loan period n
- $money_e$ currency conversion constant

Variables (positive):

- $vef_{d,e,t}$ energy flows (MWh/h)
- $vmf_{d,e,t}$ mass flows (kg/h)
- $vinvestment$ total investment costs
- $vdvcosts$ total operating costs
- $vinvest_d$ investment costs of component d
- vs_d component size
- $vl_{d,t}$ storage content

Objective

$$\max \sum_{d,e,t} p_{e,mwh} \cdot vef_{d,e,t} + \sum_{d,e,t} \pi_{e,t} \cdot vef_{d,e,t}$$

$$+ \sum_{d,e,t} p_{e,kg} \cdot vmf_{d,e,t}$$

$$- vinvestment - vdvcosts$$

Investments

$$vinvest_d = uc_d \cdot vs_d$$

$$vinvestment = \sum_n \frac{\left(\sum_{(d|n)} vinvest_d \right) * rate * (1 + rate)^n}{(1 + rate)^n - 1}$$

$$vdvcosts = \sum_n dvrates(n) * \left(\sum_{(d|n)} vinvest_d \right)$$

Balance

$$vef_{d,e,io,t} \cdot \sum_e eb_{d,io,e} = eb_{d,io,e} \cdot \sum_e vef_{d,e,io,t}$$

$$\sum_e vef_{d,e,in,t} = \sum_e vef_{d,e,out,t}$$

$$vmf_{d,e,io,t} \cdot \sum_e mb_{d,io,e} = mb_{d,io,e} \cdot \sum_e vmf_{d,e,io,t}$$

$$\sum_e vmf_{d,e,in,t} = \sum_e vmf_{d,e,out,t}$$

$$\sum_e vmf_{d,io,e,t} \cdot \sum_e eb_{d,io,e} = \sum_e vef_{d,e,io,t} \cdot \sum_e mb_{d,io,e}$$

$$vef_{SOEC,steam(free),in,t} \leq vef_{SOEC,steam,in,t} + vef_{methanator,steam(free),out,t}$$

$$vl_{H_2storage,t} + vef_{SOEC,hydrogen,out,t} = vl_{H_2storage,t+1} + vef_{methanator,hydrogen,in,t}$$

$$vl_{CH_4storage,t} + vef_{CH_4storage,CH_4,in,t} = vl_{CH_4storage,t+1} + vef_{methanator,CH_4,in,t}$$

Flow Capacity

$$vs_d = fs_d$$

$$vef_{d,e,io,t} \leq f_{d,max} \cdot eb_{d,io,e} \cdot vs_d$$

$$vef_{d,e,io,t} \geq f_{d,min} \cdot eb_{d,io,e} \cdot vs_d$$

Storage Capacity

$$vl_{d,t} \leq us_{d,e} \cdot vs_d$$

Gradients

$$vef_{d,e,io,t+1} - vef_{d,e,io,t} \leq g_{d,e}$$

$$v_{d,e,t} - v_{d,e,t+1} \leq g_{d,e}$$

.....

Results

The following analyses with the model have been made:

Configuration 1: Biogas from the biogas plant is mixed with hydrogen from the SOEC in a methanator producing SNG.

1. electricity and biogas prices for 2020 and the low SNG price for 2020
2. electricity and biogas prices for 2020 and the high SNG price for 2020
3. electricity and biogas prices for 2035 and the low SNG price for 2035
4. electricity and biogas prices for 2035 and the high SNG price for 2035

Configuration 2: (Biogas from the biogas unit is introduced directly into the SOEC unit (after purification of the biogas)).

5. electricity and biogas prices for 2020 and the low SNG price for 2020
6. electricity and biogas prices for 2020 and the high SNG price for 2020
7. electricity and biogas prices for 2035 and the low SNG price for 2035
8. electricity and biogas prices for 2035 and the high SNG price for 2035

		1	2	3	4	5	6	7	8
Electricity price	2020	X	X			X	X		
	2035			X	X			X	X
SNG price	Low	X		X		X		X	
	High		X		X		X		X
Configuration	1	X	X	X	X				
	2					X	X	X	X

Sensitivity analyses regarding investment cost for SOEC unit:

Configuration 1. Electricity and biogas prices for 2020 and the high SNG price for 2020.

9. SOEC investment * 75 %
10. SOEC investment * 150 %

Configuration 1. Electricity and biogas prices for 2035 and the high SNG price for 2035.

11. SOEC investment * 75 %
12. SOEC investment * 150 %

Sensitivity analyses regarding investment cost for hydrogen storage:

Configuration 1. Electricity and biogas prices for 2020 and the high SNG price for 2020.

13. Hydrogen storage investment * 50 %

Configuration 1. Electricity and biogas prices for 2035 and the high SNG price for 2035.

- 14. Hydrogen storage investment * 50 %
- 15. Hydrogen storage investment * 200 %

Sensitivity analyses regarding size of plant (lower specific investment for methanator and storages):

Configuration 1. Electricity and biogas prices for 2035 and the high SNG price for 2035.

- 16. Investment in methanator and storages * 60 %

Sensitivity analyses regarding investment cost for hydrogen storage and SOEC unit:

Configuration 1. Electricity and biogas prices for 2035 and the high SNG price for 2035.

- 17. Investment in hydrogen storage and SOEC unit * 50 %

		9	10	11	12	13	14	15	16	17
Based on calculation		2	2	4	4	2	4	4	4	4
SOEC inv.	%	75	150	75	150	100	100	100	100	50
Hydrogen storage inv.	%	100	100	100	100	50	50	200	60	50
Methanator inv.	%	100	100	100	100	100	100	100	60	100

Optimisation with spot prices combined with regulating power prices:

Configuration 1. Biogas prices for 2035 and the high SNG price for 2035.

- 1. Electricity prices for 2035 including supplement/reduction due to regulating power prices

7.4 Configuration 1

In the base case using configuration 1 (Biogas from the biogas plant is mixed with hydrogen from the SOEC in a methanator producing SNG), four optimisations have been made:

- 1. electricity and biogas prices for 2020 and the low SNG price for 2020
- 2. electricity and biogas prices for 2020 and the high SNG price for 2020
- 3. electricity and biogas prices for 2035 and the low SNG price for 2035
- 4. electricity and biogas prices for 2035 and the high SNG price for 2035

The optimisation of the relative sizing of the components in the system results in the same solution for calculation 1 and 2 (2020) and the same for 3 and 4 (2035). As can be seen from the table below, in 2020 the optimal solution does not include storages, whereas a seven hour hydrogen storage is included in the solution for 2035.

	2020	2035
Biogas unit	1	1
Biogas storage	0	0
SOEC	1	1,13
H2 storage	0	7
Methanator	1	1

Table 9: Relative sizing of components in calculation 1-4 (Configuration 1)

Since there is no storage in the optimal solution for 2020, the SOEC will have to be in operation all hours, and the SOEC load is therefore 100 %, as can be seen from the table below. Therefore the average power price of used power is the same as the average power for the whole year. In 2035 there is a hydrogen storage in the optimal solution, and thus the relative size of the SOEC is above 1 (1,13), and the SOEC load is 88 %.

		2020	2035
SOEC load	%	100	88
Average power price	DKK/MWh	363	347

Table 10: SOEC load and average power price of used power in the SOEC (Configuration 1)

The economic results for the four calculations with configuration 1 are shown below:

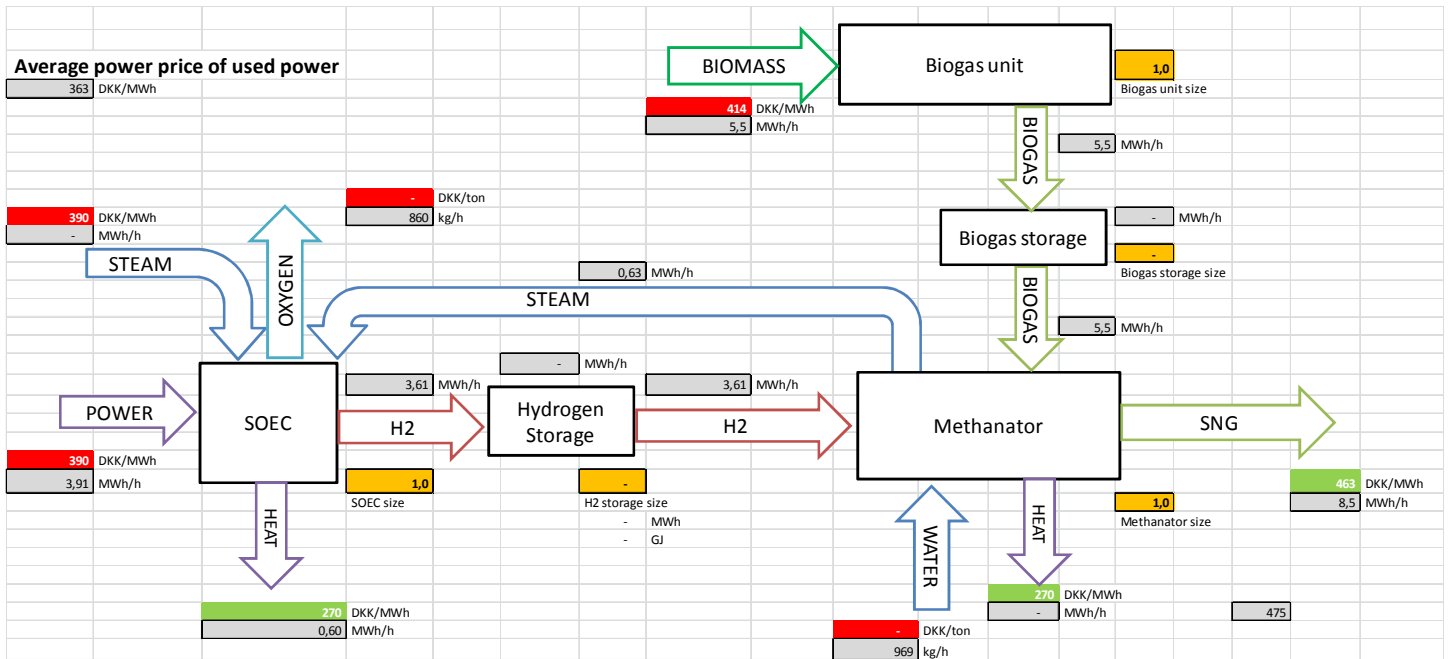
DKK per Year	2020	2020	2035	2035
	Low SNG	High SNG	Low SNG	High SNG
Added Steam (SOEC)	-	-	-117.190	-117.190
Power (SOEC)	-12.390.394	-12.390.394	-11.853.414	-11.853.414
Biogas	-20.036.540	-20.036.540	-22.998.463	-22.998.463
SOEC Inv.	-2.080.836	-2.080.836	-2.360.949	-2.360.949
Methanator Inv.	-1.604.852	-1.604.852	-1.604.852	-1.604.852
H2 Storage Inv.	-	-	-202.773	-202.773
Biogas storage Inv.	-	-	-	-
Operational cost	-1.450.447	-1.450.447	-1.637.434	-1.637.434
Total Expenses	-37.563.069	-37.563.069	-40.775.074	-40.775.074
SNG	31.441.980	37.513.083	35.997.140	42.068.243
Heat (SOEC)	1.415.232	1.415.232	1.415.232	1.415.232
Total Income	32.857.212	38.928.315	37.412.372	43.483.475
Investment year one (MDKK)	29	29	33	33
NPV	-56	19	-39	36
Profit	-4.705.857	1.365.246	-3.362.703	2.708.401

Table 11: Socio economy for configuration 1, 2020 and 2035, low/high value of SNG

As can be seen from Table 11, there is a positive profit both in 2020 and 2035, when expecting the high SNG value. When using low SNG value, there is a negative profit. The profit is higher in 2035 than in 2020, which is due to higher fluctuations in power prices and higher SNG prices.

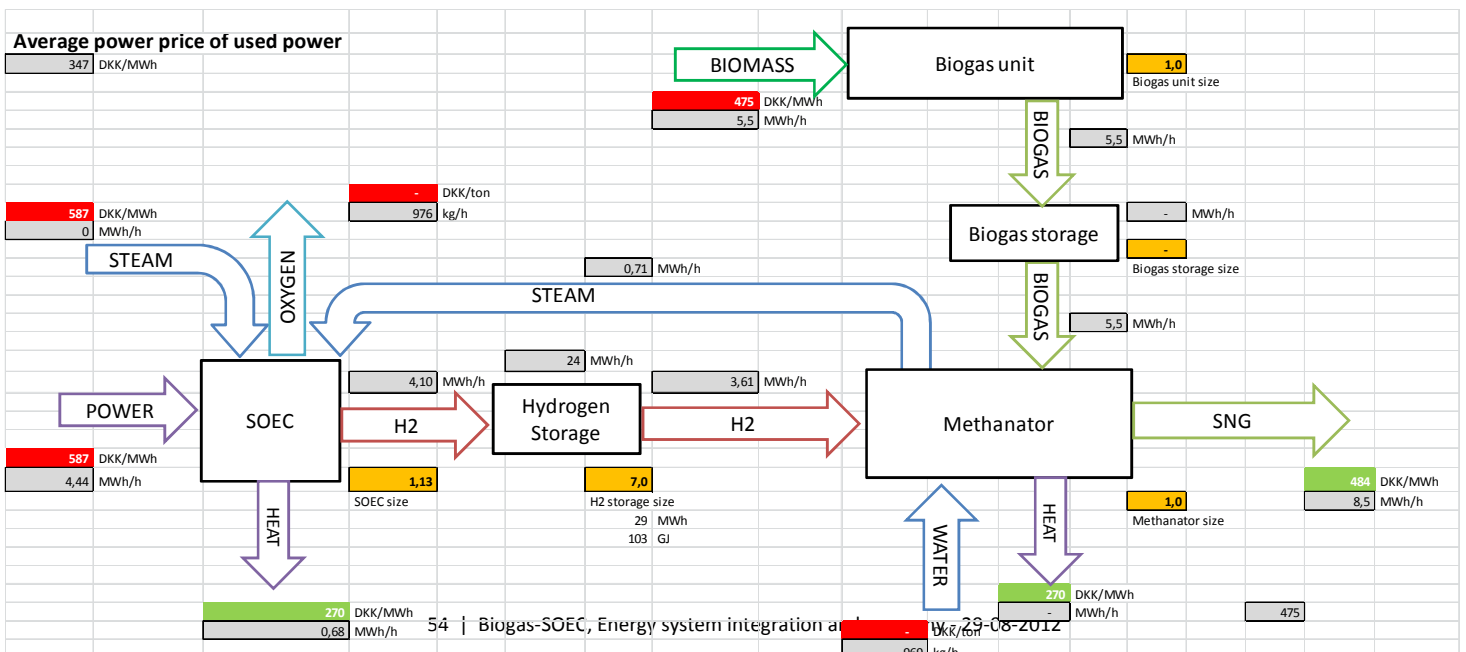
2020

In 2020 the optimal relative sizing of the components, is to have SOEC and methanator in the same size as the biogas unit. This means that no storages are necessary and that the SOEC (and methanator) is in operation every hour of the year. Hereby the average power price of used power is the same as the average power price of all hours, and the advantage of using power in the hours with the cheapest prices is lost.



2035

In 2035 the optimal relative sizing of the components are different than in 2020. Due to higher fluctuations in power prices it is more relevant to stop operation in some hours when power prices are very high. The optimal solution is with a SOEC 13 % bigger than what would be needed if the SOEC was in operation all hours. This call for a need of a storage (either hydrogen storage or biogas storage). The cheapest solution is to build a hydrogen storage rather than building a biogas storage and at the same time a bigger methanator. The optimal size of the hydrogen storage is for seven hours of hydrogen production from the SOEC.



7.5 Configuration 2

The same 4 simulations have been made using configuration 2 (Biogas from the biogas unit is introduced directly into the SOEC unit (after purification of the biogas)):

5. electricity and biogas prices for 2020 and the low SNG price for 2020
6. electricity and biogas prices for 2020 and the high SNG price for 2020
7. electricity and biogas prices for 2035 and the low SNG price for 2035
8. electricity and biogas prices for 2035 and the high SNG price for 2035

As for configuration 1 the optimal solution in 2020 for configuration 2 is a system with no storages. In 2035 there is a need for storages, to obtain an optimal solution. As opposed to configuration 1, the most profitable solution is to build a biogas storage and a small “hydrogen” storage. The “hydrogen” storage in configuration 2 is actually a syngas storage, since the biogas is introduced into the SOEC unit, and the gas from the SOEC to the “hydrogen” storage is therefore a mix mainly consisting of hydrogen, methane and carbon monoxide.

	2020	2035
Biogas unit	1	1
Biogas storage	0	7,5
SOEC	1	1,1
Syngas storage	0	1,3
Methanator	1	1,1

Table 12 Relative sizing of components in calculation 5-8 (Configuration 2)

		2020	2035
SOEC load	%	100	91
Average power price	DKK/MWh	363	350

Table 13 SOEC load and average power price of used power in the SOEC (Configuration 2)

The economic results for the 4 calculations with configuration 2 are shown below:

DKK per Year	2020	2020	2035	2035
	Low SNG	High SNG	Low SNG	High SNG
Added Steam (SOEC)	-	-	-	-
Power (SOEC)	-12.738.973	-12.738.973	-12.297.372	-12.297.372
Biogas	-20.000.373	-20.000.373	-22.956.950	-22.956.950
SOEC Inv.	-2.621.853	-2.621.853	-2.879.256	-2.879.256
Methanator Inv.	-1.283.881	-1.283.881	-1.373.306	-1.373.306
H2 Storage Inv.	-	-	-27.285	-27.285
Biogas storage Inv.	-	-	-129.871	-129.871
Operational cost	-1.367.563	-1.367.563	-1.576.931	-1.576.931
Total Expenses	-38.012.644	-38.012.644	-41.240.972	-41.240.972
SNG	31.527.836	37.689.201	36.104.381	42.265.746
Heat (SOEC)	1.485.994	1.485.994	1.485.994	1.485.994
Total Income	33.013.829	39.175.194	37.590.375	43.751.740
Investment year one (MDKK)	27	27	32	32
NPV	-60	17	-43	34
Profit	-4.998.814	1.162.551	-3.650.597	2.510.768

Table 14: Socio economy for configuration 2, 2020 and 2035, low/high value of SNG

As can be seen from Table 14, there is a positive profit both in 2020 and 2035, when expecting the high SNG value. The profit is higher in 2035 than in 2020, which is due to higher fluctuations in power prices and higher SNG prices.

The yearly profit for the optimal solutions for configuration 2 is slightly lower than for configuration 1. This is due to the smaller SOEC unit, and therefore poorer options for harvesting the lowest power prices. To increase the SOEC size in configuration 1 would mean a bigger hydrogen storage, but to increase the SOEC in configuration 2 would call for either bigger biogas storage AND syngas storage or for bigger bio gas storage AND methanator.

Since configuration 2 does not show higher profit, and the solution has more constrains regarding relative sizing of components, this configuration is not analysed further with regard to regulating power or in the sensitivity analyses.

7.6 Regulating power

The calculations above are based on buying electricity in the Nordic spot market. However, when getting closer to the actual hour of operation, other sets of electricity prices become available. These *regulating prices* represent

the value (or cost) of changing your consumption away from what was originally planned.

The regulating market

The regulating power market is an intraday market used by the transmission system operator to balance the power system within the hour of operation. Regulating power has to be activated within 15 minutes and has a stochastic nature, as the demand for regulating power arises due to failures of power plants and transmission lines as well as inaccurate forecasts for wind power production and changes in demand. For flexible units, the regulating power market offers an opportunity for additional revenues. Producers and consumers can offer bids to the transmission system operator and will be paid according to marginal pricing, if their bid is accepted. This means, the most expensive bid will set the price for all participants at a given time.

Regulating power prices

Up regulation denotes additional power production, or less power consumption, if the regulating power is delivered by a power consumer. Down regulation denotes less power production, or more power consumption, if the regulating power is delivered by a power consumer. Up regulation power prices are always higher or equal to the spot price, while down regulation power prices always are equal to or lower than the spot price. If a consumer delivers down regulation by increasing consumption, he pays a lower price for the power consumed, and saves the difference to the spot price. If a consumer stops consumption in order to deliver up regulation, he will still pay the spot price, but receive payment according to the up regulation price, and thus earn the difference.

The development of the regulating power price and the spot price in Western Denmark is shown on figure 12 for the period from January 2005 to July 2010. Compared to the monthly average spot price, up and down regulation prices were approximately 32 DKK/MWh higher and lower respectively. Figure 13 shows the percentage of the time, where regulation prices occurred. Approximately 25-30% of the time up regulation occurred and down regulation occurred approx. 25-30 % of the time as well, while no regulation prices occurred during the remaining time.

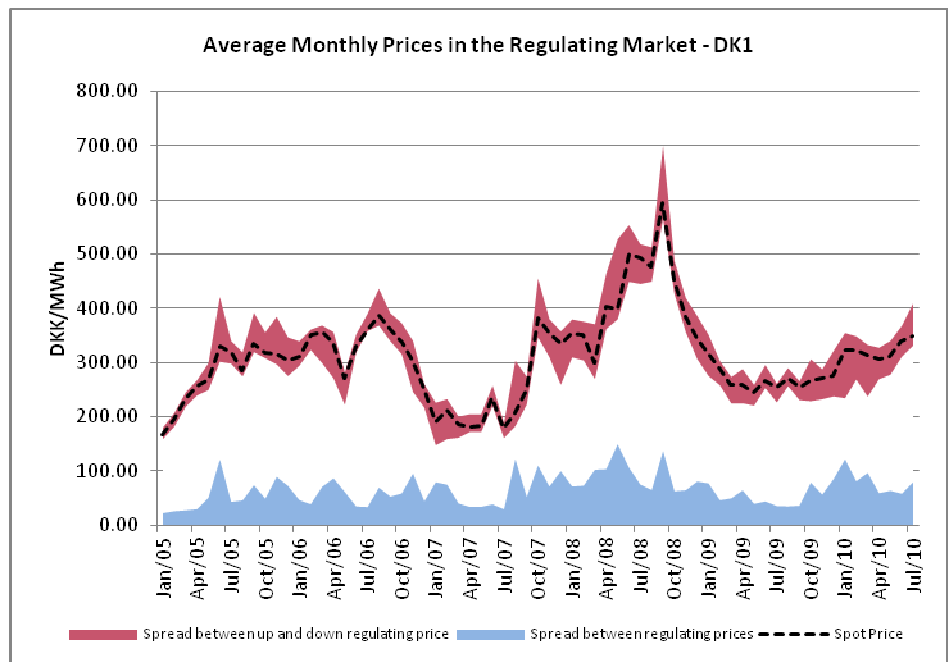


Figure 12: Average monthly spot and regulating power prices for DK 1 from January 2005 to July 2010. The dotted black line is the spot price, while the red portion represents the up and down regulating prices. This spread is also displayed in blue at the bottom of the figure. Note that this is monthly average. Hourly values vary much more. Source: The existing Nordic regulating power market, FlexPower WP1 – Report 1, Ea energy Analysis, May 2012

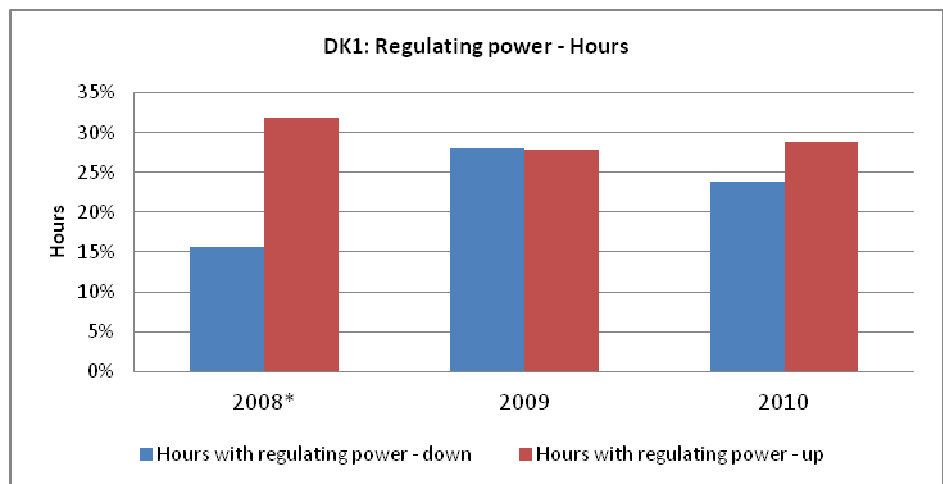


Figure 13: Percentage of total hours with regulating power activated in DK1 and DK2 from June 1st, 2008 till August 10th, 2010. *Indicates data started on June 1st. Source: The existing Nordic regulating power market, FlexPower WP1 – Report 1, Ea Energy Analysis, May 2012

The regulating power market in Denmark is supplemented with a reservation market, which aims at securing sufficient amounts of regulating power during the hour of operation. If a unit receives a reservation price, it has to be available for up/down regulation during the operating hour. Actual activation will be paid separately according to the regulating power price.

Development of regulating power market

The development of the regulating power market is hard to predict. An assessment carried out by Ea Energy Analysis for a project on combined wind power and heat pumps⁵ pointed out a number of factors influencing the future demand of regulating power. While increasing amounts of wind power and fewer central power stations will increase the demand for regulating power, other factors, such as better market integration with neighbouring countries and increased transmission capacities will limit the need for regulating power. Overall the estimate pointed out, that the level of regulating power prices will not change significantly, also because more suppliers will enter the market and increase the possible supply of regulating power.

FlexPower⁶ is a research project examining the potential for activating demand as regulating power. As part of the project a Markov chain model for generation of regulating power prices was developed. This model can be used to generate a time series of regulating power prices. A Markov chain describes a time series based on probabilities of going from one state to another. Market data for regulating power prices (calculated as difference from the spot price) from 2002 to 2009 were categorised into intervals of 100 DKK/MWh and based on the statistics the probability of going from one interval to another was calculated. This is shown on table 15. The high values in the diagonal indicate a high probability that the price during the next hour is similar to the current hour.

	-4	-3	-2	-1	0	1	2	3	4
-4	30%	17%	10%	12%	13%	2%	0%	0%	0%
-3	5%	51%	18%	8%	13%	2%	0%	0%	0%
-2	1%	6%	51%	21%	16%	3%	0%	0%	0%
-1	0%	1%	4%	66%	24%	4%	0%	0%	0%
0	0%	0%	1%	5%	88%	5%	1%	0%	0%
1	0%	0%	1%	4%	25%	64%	4%	1%	0%
2	0%	0%	0%	1%	20%	25%	41%	8%	2%
3	0%	0%	1%	1%	21%	13%	22%	31%	5%
4	0%	0%	0%	1%	21%	12%	8%	14%	26%

Table 15: The centre part of the Markov matrix. If the current state is 0 (value of regulating power minus spot price is between 0 and 100 DKK/MWh), the probability for staying in this state is 88%, and the probability of going to state +1 (100-200 DKK/MWh) or -1 (-100-0 DKK/MWh) is both 5%. Source: FlexPower project.

⁵ Kombination af vindkraft og varmepumpe til Varmeplan Århus, Ea Energianalyse, Vind Energi Danmark, Nordjysk Elhandel and AffaldVarme Århus, march 2010

⁶ http://www.ea-energianalyse.dk/projects-danish/1027_flexpower_markedsdesign.html

The Markov chain model from the FlexPower project is used here to simulate a time series showing the difference between the spot price and the regulating power price. A duration curve for the developed price curve is shown on Figure 14, showing a higher variation for the regulation power price, with both higher and lower values. The variations of the simulated time series for the regulating power price are independent of the variations and level of the spot price and depend only on the simulated difference between spot and regulating power price from the previous hour. This is a limitation of the simulated series used here, as the difference between the regulating power price and the spot price in reality most likely will show some dependence on the spot price level. Furthermore, the simulation assumes unchanged characteristics of the regulating power price compared to the period used for estimating the Markov chain model. This is true both for the level and the variations of the regulating power price.

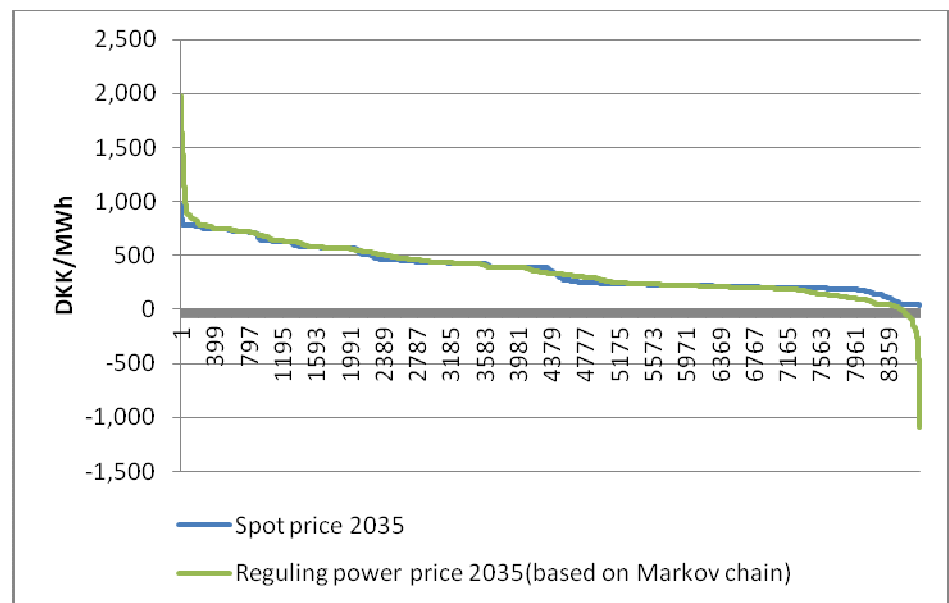


Figure 14: Simulated spot price and regulating power price.

Regulating power market potential for Biogas-SOEC

The economic potential of using the regulating power market is analysed by using the developed time series for regulating power prices described above. This is done by feeding the new time series into the model (regsim), and comparing the results to the simulation with the spot prices (spotsim) for the year 2035. The size of the components is kept constant at the level estimated during spotsim and thus only the production pattern is changed. Possible earnings from reservation payments are not considered.

The approach for calculating revenues from using the regulating power market described below aims at giving an estimate of the optimal earnings and does not represent a practical strategy.

The regulating power prices will give the SOEC-unit the opportunity to use the low down regulation price. However, in practise, this requires full foresight, since the unit has to stay out of the spot market and wait for the lower down regulation price. At the same time the unit is forced to produce at some higher prices, at times where the regulation power price is high, and electricity consumption cannot be avoided due to storage constraints. If full foresight is assumed here as well, the SOEC-unit will not pay the up regulation price, but only the spot price, as consumption is planned for the spot market already. Finally the SOEC-unit has the option to stop consumption in order to deliver up regulation. This is not directly represented as an incentive in the model, but can to some extent be calculated afterwards. To capture these aspects, the economy by using regulating power prices is analysed in the following steps:

1. Electricity payment by using spot prices only
2. Electricity payment by using regulating power prices only (spot price if difference between spot and regulating power price is zero)
3. Electricity payment by optimal usage of down regulation prices and spot prices (using down regulation price where possible and paying spot price at other times)
4. Net electricity payment by optimal usage of down regulation prices and spot prices and additional earnings from up regulation. Up regulation profit is estimated by calculating the profit during the hours, where the SOEC-unit is running during spotsim but stopped during regsim.

	Net electricity payment	Savings rel. to spotsim
	Mio DKK/year	Mio DKK/year
1 Spot prices only	11,8	-
2 Regulating power prices only	11,3	0,58
3 Optimal use of spot price and down regulation price	10,8	1,0
4 Optimal use of spot price and down regulation price and earnings from up-regulation.	10,4	1,5

Table 16: Net electricity payments by using the regulating power market. The total annual power consumption is the same for all cases.

The calculations show total savings of approx. 1.5 mill. DKK (13 %) compared to the simulation based on spot prices only. Approximately 1 mill. DKK are based on a perfect forecast of down regulation prices, while approx. 0.5 mill. are based on earnings from up regulation.

The above estimates of savings from using the spot market are optimistic, since full foresight requires perfect forecast models. The savings from using the down regulation price can be hard to realise in practice, especially if the SOEC-unit has a high number of required operating hours.

On the other hand, earnings from stopping the unit might be underestimated. This is because the model can only see the incentive of avoiding an expensive hour and using power during a cheaper hour. It does not receive the extra payment arising from stopping the unit. Therefore, the usage of up regulation prices is not optimised, but earnings are instead estimated after the simulation. If the incentive was implemented in the model, there would be a larger incentive to change the production pattern compared to the spotsim-case, thus increasing the earnings from delivering up regulation. As the unit has a high number of operating hours, up regulation is likely to be the most interesting market for the SOEC-unit in practice.

A simple strategy for using the regulating power market could be to plan consumption according to the spot market. Afterwards, the unit can offer up or down regulation depending on the operational state from the spot market plan. For units with a fixed threshold price (electricity price under which electricity consumption is beneficial), this strategy does not require advanced forecasts. However, since the SOEC-unit has to plan electricity consumption depending on different storage constraints, at least a forecast for spot market

prices will be necessary, regardless of whether opportunities in the regulating power market are utilised or not.

Based on the simple analysis carried out here, proper usage of regulating power prices can reduce the annual electricity payments by approximately 10 % if full foresight is available. A more realistic level might be around 5 %.

Finally reservation payments can potentially increase the total earnings from the regulating power market. However, the reservation market will restrict the options for the spot market. E.g. if the unit receives reservation payment for up regulation, the unit has to be running in the spot market in order to be able to deliver up regulation - regardless of the final spot price.

7.7 Sensitivity analysis

Sensitivity analyses have been made regarding investment costs for the SOEC unit, investment costs for the hydrogen storage and size of the plant. The sensitivity analyses are made based on the base case, which is configuration 1 with high SNG price and other prices from either 2020 or 2035.

SOEC investment cost

There is a high uncertainty regarding the level of the investment cost for a large scale SOEC unit in the future. Therefore sensitivity analyses have been made regarding investment cost for the SOEC unit. The calculations are made based on configuration 1 using electricity and biogas prices for 2020 and the high SNG price for 2020. The investment costs have in two optimisations been reduced with 25 % and increased with 50 %.

9. 2020. SOEC investment * 75 %

10. 2020. SOEC investment * 150 %

The relative sizing of the components do not change compared to the base case, meaning that the optimal solution in both cases are without any storages.

The economic results for the sensitivity calculations are in the table below compared to the base case:

DKK per Year	Base: 2020 High SNG	SOEC inv*1,5	SOECinv*0,75
Added Steam (SOEC)	-	-	-
Power (SOEC)	-12.390.394	-12.390.394	-12.390.394
Biogas	-20.036.540	-20.036.540	-20.036.540
SOEC Inv.	-2.080.836	-3.121.254	-1.560.627
Methanator Inv.	-1.604.852	-1.604.852	-1.604.852
H2 Storage Inv.	-	-	-
Biogas storage Inv.	-	-	-
Operational cost	-1.450.447	-1.675.670	-1.337.835
Total Expenses	-37.563.069	-38.828.710	-36.930.248
SNG	37.513.083	37.513.083	37.513.083
Heat (SOEC)	1.415.232	1.415.232	1.415.232
Total Income	38.928.315	38.928.315	38.928.315
Investment year one (MDKK)	29	34	27
NPV (MDKK)	19	4	27
Profit	1.365.246	99.605	1.998.067

Table 17: Socio economy for 2020 high value of SNG, sensitivity analyses regarding investment in SOEC compared to base case.

As can be seen from the table above, the only thing changing is the investment cost in the SOEC itself (and the operational cost, since these are calculated as a fraction of total investment costs). In 2020 the investment cost will not influence the optimal relative sizing of the plant, nor the operational strategy. The change in investment cost will affect the yearly profit proportionate to the change in investment level.

To see the effect of changing the SOEC investment price on the storage optimisation, the sensitivity analyses are also made with 2035 as base case:

11. 2035. SOEC investment * 75 %
12. 2035. SOEC investment * 150 %

Relative sizing of components:

	Base: 2035 High SNG	SOEC inv*1,5	SOEC inv*0,75
Biogas unit	1	1	1
Biogas storage	0	1,6	0
SOEC	1,1	1	1,2
H2 storage	7	0,2	10,9
Methanator	1	1	1

Table 18: Relative sizing of components for 2035 high value of SNG. Sensitivity analyses regarding investment in SOEC unit compared to base case.

As can be seen in the table above, a cheaper SOEC unit will result in building a bigger SOEC unit and also a bigger hydrogen storage, whereas a more expensive SOEC storage will result in building a smaller SOEC.

		Base: 2035 High SNG	SOEC inv*1,5	SOEC inv*0,75
SOEC load	%	88	98	81
Average power price	DKK/MWh	347	368	333

Table 19: SOEC load and average power price of used power in the SOEC. Based on 2035 high value of SNG. Sensitivity analyses regarding investment in SOEC unit compared to base case.

Only changing the SOEC investment cost has surprisingly little impact on the SOEC load. It would be expected that a cheaper SOEC unit would result in a solution with a much bigger SOEC unit in order to harvest more of the low electricity price hours. Even with a 25 % reduction of the SOEC investment cost, the optimal SOEC load is still rather high (81 %).

To reduce the SOEC load significant, several parameters must be changed:

- the SOEC investment cost must be lower
- the hydrogen storage investment cost must be lower
- perhaps the variation in electricity prices must also be higher

The economic results for the sensitivity calculations are in the table below compared to the base case for 2035:

DKK per Year	Base: 2035 High SNG	SOEC inv*1,5	SOECinv*0,75
Added Steam (SOEC)	-117.190	-3.331	-179.258
Power (SOEC)	-11.853.414	-12.554.426	-11.381.695
Biogas	-22.998.463	-22.998.463	-22.998.463
SOEC Inv.	-2.360.949	-3.180.260	-1.932.205
Methanator Inv.	-1.604.852	-1.630.175	-1.604.852
H2 Storage Inv.	-202.773	-5.885	-315.884
Biogas storage Inv.	-	-27.711	-
Operational cost	-1.637.434	-1.725.156	-1.615.103
Total Expenses	-40.775.074	-42.125.407	-40.027.461
SNG	42.068.243	42.068.243	42.068.243
Heat (SOEC)	1.415.232	1.415.232	1.415.232
Total Income	43.483.475	43.483.475	43.483.475
Investment year one (MDKK)	33	35	32
NPV (MDKK)	36	20	45
Profit	2.708.401	1.358.068	3.456.014

Table 20 Socio economy for 2035 high value of SNG, sensitivity analyses regarding investment in the SOEC unit compared to base case.

As can be seen from the economic comparison, the reduction in electricity cost is not very high going from the base case to the case with 25 % cheaper SOEC unit, even though the SOEC load is reduced from 88 to 81 %. This suggests that the variation in electricity prices is not very high, when looking at less than 24 hours.

As expected the economy will profit from a cheaper SOEC unit. The positive result is that even with a 50 % more expensive SOEC unit, there is a positive yearly profit.

Hydrogen storage investment costs

There are not many larger hydrogen storage facilities, and the information regarding investment costs are scarce. This fact introduces a high uncertainty regarding the level of the investment cost for a hydrogen storage in these calculations. Therefore sensitivity analyses have been made regarding investment cost for the hydrogen storage.

The calculations were initially made based on configuration 1 using electricity and biogas prices for 2020 and the high SNG price for 2020 (base case).

13. 2020. Hydrogen storage investment * 50 %

In the base case in 2020 the optimal solution does not include hydrogen storage. Even if the investment cost in the hydrogen storage was only the half, the optimal solution still does not include hydrogen storage. The optimal solution and also the economy are the same for the two cases:

- 2020, high SNG price, investment in hydrogen storage 100 DKK/kWh
- 2020, high SNG price, investment in hydrogen storage 50 DKK/kWh

Therefore the sensitivity analyses for the investment in the hydrogen storage is made for the 2035 base case (with high SNG price). Two optimisations are made:

14. 2035. Hydrogen storage investment * 50 %
15. 2035. Hydrogen storage investment * 200 %

As one would expect, the optimal solution include a bigger hydrogen storage (as can be seen in the table below), if the investment costs for the storage are lowered to the half. If, on the other hand, the investment costs are doubled, it's more profitable to build a storage for the biogas and increase the size of the methanator.

Relative sizing of components:

	Base: 2035 High SNG	H2 sto inv: 50 %	H2 sto inv: 200 %
Biogas unit	1	1	1
Biogas storage	0	0	5,9
SOEC	1,1	1,3	1,1
H2 storage	7	20,8	0
Methanator	1	1	1,1

Table 21: Relative sizing of components for 2035 high value of SNG, sensitivity analyses regarding investment in hydrogen storage compared to base case.

		Base: 2035 High SNG	H2 sto inv: 50 %	H2 sto inv: 200 %
SOEC load	%	88	77	94
Average power price	DKK/MWh	347	318	356

Table 22: SOEC load and average power price of used power in the SOEC. Based on 2035 high value of SNG. Sensitivity analyses regarding investment in hydrogen storage compared to base case.

The economic results for the sensitivity calculations are in the table below compared to the base case:

DKK per Year	Base: 2035 High SNG	H2 sto inv: 50 %	H2 sto inv: 200 %
Added Steam (SOEC)	-117.190	-242.499	-
Power (SOEC)	-11.853.414	-10.851.827	-12.153.449
Biogas	-22.998.463	-22.998.463	-22.998.463
SOEC Inv.	-2.360.949	-2.706.928	-2.224.793
Methanator Inv.	-1.604.852	-1.604.852	-1.715.879
H2 Storage Inv.	-202.773	-300.699	-
Biogas storage Inv.	-	-	-102.140
Operational cost	-1.637.434	-1.773.348	-1.614.436
Total Expenses	-40.775.074	-40.478.616	-40.809.161
SNG	42.068.243	42.068.243	42.068.243
Heat (SOEC)	1.415.232	1.415.232	1.415.232
Total Income	43.483.475	43.483.475	43.483.475
Investment year one (MDKK)	33	35	32
NPV (MDKK)	36	40	36
Profit	2.708.401	3.004.859	2.674.314

Table 23: Socio economy. Based on 2035 high value of SNG. Sensitivity analyses regarding investment in hydrogen storage compared to base case.

The yearly profit when reducing the investment cost for the hydrogen storage is obviously higher than in the base case. The yearly profit when increasing the investment cost for the hydrogen storage is close to the base case. This is because the optimal solution using a biogas storage or using a hydrogen storage are quite close to each other with the investment cost used in the base case.

Bigger plant

A sensitivity analyses is made to evaluate the effect of “economy by scale”. If the Biogas-SOEC plant was 4 times bigger than in the case studied in this report, what would be the effect?

For a 4 times bigger plant it is expected that the investment in the SOEC unit is linear scaled. There is not much to save by building a bigger SOEC, since the majority of the investment is in the cells. Other investments (in hydrogen storage, biogas storage and methanator) are on the other hand expected to have cheaper specific investment, when scaling up. If 4 times bigger, the investments are only expected to be $4 * 0,75^2$ higher.

The sensitivity analyses regarding size of plant is based on configuration 1, with electricity and biogas prices for 2035 and the high SNG price for 2035.

16. Investment in methanator and storages * 60 %

When building a bigger plant and hereby reducing the specific investment in methanator and storages, the optimal solution favours increasing the size of the biogas storage and the methanator. The SOEC unit is also slightly larger than in the base case to match the bigger methanator.

Relative sizing of components:

	Base: 2035 High SNG	4* bigger
Biogas unit	1	1 (4)
Biogas storage	0	16,7 (66,8)
SOEC	1,1	1,2 (4,8)
H2 storage	7	4 (6)
Methanator	1	1,2 (4,8)

Table 24: Relative sizing of components for 2035 high value of SNG, sensitivity analyses regarding 4 times bigger plant compared to base case.

The bigger SOEC unit results in lower average price of the used power, as can be seen in the table below.

		Base: 2035 High SNG	4* bigger
SOEC load	%	88	80
Average power price	DKK/MWh	347	323

Table 25: SOEC load and average power price of used power in the SOEC. Based on 2035 high value of SNG. Sensitivity analyses regarding 4 times bigger plant compared to base case.

The economic results for the sensitivity calculations are in the table below compared to the base case:

DKK per Year	Base: 2035 High SNG	4* bigger divided by 4 for comparing to base case	4* bigger
Added Steam (SOEC)	-117.190	-52.310	-209.241
Power (SOEC)	-11.853.414	-11.020.750	-44.083.000
Biogas	-22.998.463	-22.998.463	-91.993.854
SOEC Inv.	-2.360.949	-2.591.669	-10.366.677
Methanator Inv.	-1.604.852	-1.135.983	-4.543.932
H2 Storage Inv.	-202.773	-69.715	-278.861
Biogas storage Inv.	-	-174.740	-698.960
Operational cost	-1.637.434	-1.421.194	-5.684.777
Total Expenses	-40.775.074	-39.464.825	-157.859.301
SNG	42.068.243	42.068.243	168.272.972
Heat (SOEC)	1.415.232	1.415.232	5.660.928
Total Income	43.483.475	43.483.475	173.933.900
Investment year one (MDKK)	33	28	114
NPV (MDKK)	36	52	210
Profit	2.708.401	4.018.650	16.074.599

Table 26: Socio economy. Based on 2035 high value of SNG. Sensitivity analyses regarding 4 times bigger plant compared to base case.

The yearly profit for a 4 times bigger plant is, not surprisingly, more than 4 times higher than the base case. Scaled to the same size the yearly profit or the NPV is almost 50 % higher for the bigger plant. In these calculations potential higher cost for the biogas due to longer transportation of the manure is not taken into account.

Lower investment costs for SOEC unit and hydrogen storage

In the last sensitivity analyses made, the investment level in the SOEC unit and the hydrogen storage is reduced to 50 % to see if this will result in more dynamic operation for the SOEC unit.

The sensitivity analyses regarding investment cost for hydrogen storage and SOEC unit is based on configuration 1, with electricity and biogas prices for 2035 and the high SNG price for 2035.

17. Investment in hydrogen storage and SOEC unit * 50 %

With lower investment level for hydrogen storage and SOEC unit, the optimal solution favours increasing the size of the hydrogen storage and the SOEC unit. This leads to more dynamic operation of the SOEC unit.

Relative sizing of components:

	Base: 2035 High SNG	H2 sto and SOEC inv: 50 %
Biogas unit	1	1
Biogas storage	0	0
SOEC	1,1	1,8
H2 storage	7	40
Methanator	1	1

Table 27: Relative sizing of components for 2035 high value of SNG, sensitivity analyses regarding lower investment in SOEC unit and hydrogen storage compared to base case.

As expected the lower investment costs for the SOEC unit and the hydrogen storage will result in building a bigger SOEC unit and hydrogen storage.

The bigger SOEC unit and hydrogen storage results in much lower average price of the used power, because the SOEC will only be in operation about half of the hours of the year.

		Base: 2035 High SNG	H2 sto and SOEC inv: 50 %
SOEC load	%	88	55
Average power price	DKK/MWh	347	273

Table 28: SOEC load and average power price of used power in the SOEC. Based on 2035 high value of SNG. Sensitivity analyses regarding lower investment in SOEC unit and hydrogen storage compared to base case.

The optimal solution call for a SOEC unit only 1,8 times bigger than the methanator, but a hydrogen storage for 40 hours of operation of the methanator. This indicates that the hours with low electricity prices are not evenly distributed over the year.

The economic results for the sensitivity calculations are in the table below compared to the base case:

DKK per Year	Base: 2035 High SNG	H2 sto and SOEC inv: 50 %
Added Steam (SOEC)	-117.190	-451.250
Power (SOEC)	-11.853.414	-9.336.531
Biogas	-22.998.463	-22.998.463
SOEC Inv.	-2.360.949	-1.899.176
Methanator Inv.	-1.604.852	-1.604.852
H2 Storage Inv.	-202.773	-573.834
Biogas storage Inv.	-	-
Operational cost	-1.637.434	-1.768.684
Total Expenses	-40.775.074	-38.632.789
SNG	42.068.243	42.068.243
Heat (SOEC)	1.415.232	1.874.686
Total Income	43.483.475	43.942.929
Investment year one (MDKK)	33	35
NPV (MDKK)	36	69
Profit	2.708.401	5.310.140

Table 29: Socio economy. Based on 2035 high value of SNG. Sensitivity analyses regarding lower investment in SOEC unit and hydrogen storage compared to base case.

The yearly profit is doubled in this sensitivity calculation compared to the base case. Half of this increase is directly due to the lower investment costs the other half is due to the lower investment cost enabling bigger SOEC unit and hydrogen storage and hereby enabling operation in hours with lower electricity prices.

Reference list

- Danish Energy Agency: "Anvendelse af biogasressourcerne og gasstrategi herfor", May 2010
- Danish Energy Agency and Energinet.dk: "Technology data for Energy Plants", June 2010
- RUC: "Scenarier for samlet udnyttelse af brint som energibærer i Danmarks fremtidige energisystem, Bilag A: Teknologikatalog", 2001
- Ea Energy analyses: "Biomassehandlingsplaner for Randers, Norddjurs og Syddjurs, Enercoast slutkonference", 27. June 2012
- Technical information from John Bøggild Hansen, Claus Friis Pedersen and Troels Stummann from Haldor Topsøe, 2011-2012
- Ea Energy analyses: "The existing Nordic regulating power market, FlexPower WP1 – Report 1", May 2012
- Ea Energianalyse, Vind Energi Danmark, Nordjysk Elhandel and AffaldVarme Århus: "Kombination af vindkraft og varmepumpe til Varmeplan Århus", March 2010
- http://www.ea-energianalyse.dk/projects-danish/1027_flexpower_markedsdesign.html
- FlexPower project
- EU Commission: Roadmap for moving to a competitive low carbon economy in 2050, 2011
- EU Commission: "EU energy Trends 2030", 2011
- IEA: "World Energy Outlook 2011", 2011
- UN, "Copenhagen Accord", 2009
- Danish Government Platform: "Et Danmark, der står sammen", 2011
- Danish Commission On Climate Change Policy: "Green Energy – the road to a Danish energy system without fossil fuels", 2010
- The Danish Government: "Energy agreement", March 2012
- PlanEnergi: "Biogaspotentiale i danske kommuner", May 2011
- Danish Energy Agency: "Forudsætninger for samfundsøkonomiske analyser på energiområdet", April 2011

Appendix A: Subsidies for biogas

In the following existing subsidies and taxes for biogas is reviewed.

Electricity price premium	<p>The existing subsidy scheme provides all new and existing biogas plants with a fixed electricity price of 745 DKK/MWh or a fixed price premium of 405 DKK/MWh (2008 prices) when biogas is used in relation with other fuels. These subsidies are regulated in relation to 60 % of the yearly increase in the net price index. With this price index the current electricity price premium is 424 DKK/MWh.</p> <p>Besides biogas this subsidy also covers electricity generated by gasification using biomass, sterling engines or other special electricity plants using biomass.</p>
Tax exemption	<p>Heat generated using biogas is exempted from heat taxes. When biogas displaces natural gas CHP this corresponds to an indirect subsidy of 55.6 kr./GJ.</p>
Taxes on Methane tax	<p>On the 1st of January 2011 a methane tax was introduced on natural gas and biogas used as a fuel in motors plants. Biogas or natural gas used in gas turbines or heat boilers is not covered by this tax. This tax is 1.6 DKK/GJ on natural gas and 1.1 DKK/GJ on biogas. For natural gas this tax is increased by 1.8 % yearly until 2015 and is hereafter set to follow the price in the net price index.</p>
NOx tax	<p>Since 2010 a tax on fuels emitting NOx in combustion has been in force. When measuring the NOx emission the tax is 5.20 DKK/kg NOx. From the 1st of July 2012 this tax is increased to 25 DKK/kg. If no measuring is taking place a payment based on a set of standards are made.</p>
CO2 tax, transport	<p>Biogas is not imposed with the CO2 tax unless it is used for transport.</p>
Security of supply tax	<p>In the energy agreement biogas is also set to be imposed with a security of supply tax. This tax will be in force from 2013 and the exact outline of this is not yet set, but we expect that in 2020 the tax will amount to approx. 27 DKK/GJ for bioenergy and approx. 20 DKK/GJ for fossil fuels.</p>

New incentives included in the March 2012 agreement

Biogas used for CHP or in the natural gas grid will receive a subsidy of approximately 115 DKK/GJ from 2012 through a new approach for biogas subsidy. This approach includes:

- Biogas for CHP and biogas delivered to the natural gas grid will have equal status meaning that also biogas delivered to the natural gas grid will receive the base subsidy of 79 DKK /GJ.
- A new base subsidy is introduced for biogas used for transport or process industry of 39 DKK /GJ.
- The subsidy for construction of the biogas plant will be increased from 20 to 30 %. This subsidy can also include the investments needed on the farms in relation to the biogas plant.
- A new subsidy of 26 DKK /GJ is introduced for all biogas usage. The subsidy will decrease in relation with the increase in natural gas prices. The subsidy will decrease with 0.01 DKK/GJ when the natural gas price increases 0.01 DKK/GJ. This subsidy will not cover biogas produced on e.g. maize.
- An additional subsidy of 10 DKK /GJ for all usage of biogas is also introduced. The subsidy will decrease by 2 DKK /GJ a year from 2016 to 0 DKK /GJ in 2020.

Appendix B: Energy and CO2 taxes

The total energy and CO2 taxes for heat production are shown in the table below.

(2011-prices)	2011	2015	2025
Coal	61.2	64.7	69.1
Natural gas	56.2	59.7	64.1
Fuel oil	59.3	62.8	67.2
Gas oil	58.3	61.9	66.3
Waste	54.3	57.8	62.2

Table 1: Energy and CO2 taxes, total. (DKK/GJ)

Electricity is taxed on the consumption and heat on the input side. For fuels used for CHP an artificial heat efficiency of 120 % is applied. In practice this means that the tax is divided by 1.2 when the fuels are used for CHP.

Part 3

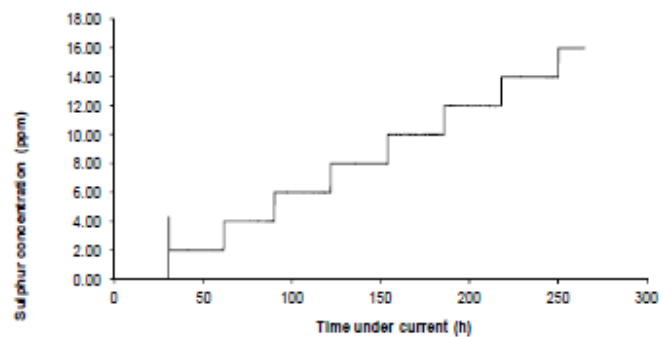
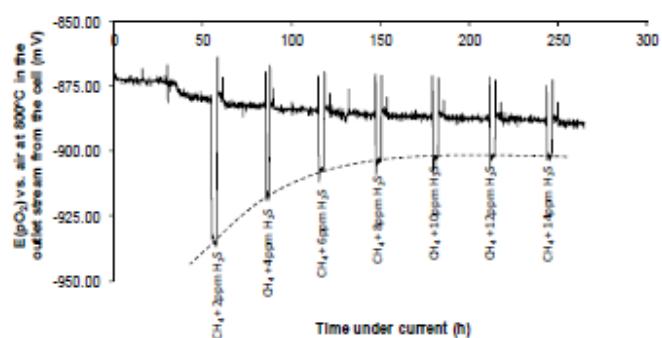
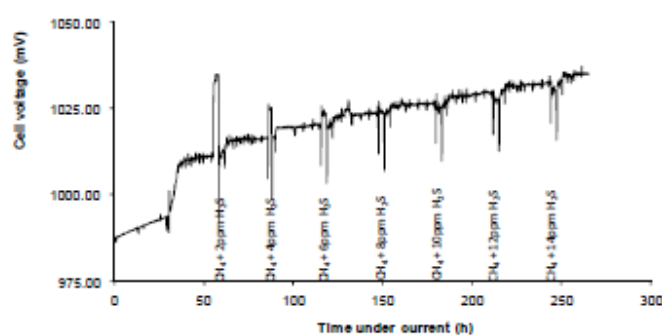
Biogas upgrading using SOEC with Ni-ScYSZ electrode

Sune D. Ebbesen, Mogens B. Mogensen

Department of Energy Conversion and Storage

Technical University of Denmark, DTU,

Frederiksborgvej 399, DK-4000 Roskilde



1 Introduction

The purpose of this part of the project is to investigate experimentally the feasibility of electrochemical upgrading of biogas by converting the carbon dioxide and steam in the biogas to a mixture of carbon monoxide and hydrogen by Solid Oxide Electrolysis Cells (SOECs). By supplying a methane containing biogas to the SOEC directly, methane will be converted to carbon monoxide and hydrogen (steam reforming) which is disadvantageous. The possibility of leaving some sulphur in the biogas feed to the SOEC, to reduce the steam reforming activity without sacrificing too much of the electrochemical electrolysis activity is investigated. Since scandia containing SOCs has shown better tolerance towards sulphur impurities¹ when operated as fuel cells, the SOCs applied for this study is based on Ni-ScYSZ (Ni-ScYSZ/ScYSZ/CGO_{barrier}-LSC-CGO). This report describes all experiments performed at DTU Energy conversion (former Risø DTU).

2 Experimental

Two planar Ni-ScYSZ-supported SOCs of 5×5 cm² with an active electrode area of 4×4 cm² - supplied by Topsoe Fuel Cell as part of their work package - were used for the experiments. At start-up, the nickel oxide in the Ni-YSZ electrode is reduced to nickel in hydrogen at 1000 °C. Further one metal supported cell produced at DTU Energy conversion was tested without sulphur in the gas. This cell has a fuel electrode that consists of porous Fe – 22% Cr-based stainless steel alloy with up to 50 vol% doped zirconia electrolyte as a backbone structure, which is infiltrated with a solution comprising precursors for Ce_{0.8}Gd_{0.2}O_{1.9} (CGO20) + 10 wt.% Ni (with respect to CGO20) to form the active fuel electrode. After infiltration, the cells were calcined at 350 °C. The low Ni content is believed to make this cell very suitable for biogas upgrading. Further cell information can be found elsewhere^{2,3}. The cell assembly used for these experiments have been described in detail elsewhere^{4,5}.

2.1 Initial Electrochemical Characterisation

After reduction, the cell was characterised in H₂O – H₂ mixtures following a standard procedure at DTU Energy conversion. This procedure consists of AC and DC characterisation in the temperature range from 750 °C to 850 °C with various gas mixtures supplied to the Ni-YSZ electrode (4 % H₂O – 96 % H₂, 20 % H₂O – 80 % H₂, 50 % H₂O – 50 % H₂), and pure oxygen or air supplied to the Oxygen electrode. Further, additional AC and DC characterisation was performed in simulated biogas supplied to the Ni-ScYSZ electrode.

The DC characterisation of the cell was performed by recording polarisation curves (i-V curves) in both electrolysis and fuel cell mode by varying the current. AC characterisation was performed by Electrochemical Impedance Spectroscopy (EIS) using an external shunt and a Solartron 1255B or 1260 frequency analyzer at frequencies from 82 kHz to 0.08 Hz. The impedance data were corrected using the short-circuit impedance response of the test set-up. From the impedance spectra, the ohmic (serial) resistance (R_s) is taken as the value of the real part of the corrected impedance measured at 82 kHz. The electrochemical polarisation resistance means here the polarisation resistance minus the concentration resistance. The polarisation resistance (R_p) is taken as the difference in real part of the impedance at 82 kHz and 0.08 Hz. The total Area Specific Resistance (ASR) of a cell is calculated as the total AC resistance of the real part ($R_s + R_p$, to 0.08 Hz) of the impedance.

2.2 Durability of the Solid Oxide Electrolysis Cells

The durability of the SOCs during electrolysis of simulated biogas was examined using two identical cells; both operated at 800 °C. The durability test of the first cells was intended as a reference test, and operated on simulated biogas where methane was substituted with nitrogen (to simulate the gas mixture where steam reforming is suppressed, 51.4% H₂O, 5.1% H₂, 1.6% CO, 15.8% CO₂, and 26.1% N₂) and without the addition of sulphur. The second test was a number of experiments with the addition of both methane and sulphur to examine the effect of sulphur on the steam reforming activity (the gas compositions supplied to the Ni-ScYSZ electrode are shown in Table 1).

2.2.1 AC- Characterisation during Durability Testing

Electrochemical impedance spectra were also recorded during the electrolysis tests to examine the detailed behaviour of the cells. To improve the frequency resolution of the spectra recorded during electrolysis testing, Analysis of the Difference in Impedance Spectra (ADIS) was performed⁶. The difference in the impedance was calculated from the real part of the experimental impedance, $Z'(f)$, according to equation (I) with $Z'(f)_{t=\text{reference time}}$ used as the reference. The reference time is either the start of electrolysis or when sulphur is introduced. The specific reference time will be stated in the text.

$$\frac{\Delta_t \partial Z'(f)}{\partial \ln(f)} = \frac{\left(Z'(f_{n+1})_t - Z'(f_{n-1})_t \right) - \left(Z'(f_{n+1})_{t=\text{reference time}} - Z'(f_{n-1})_{t=\text{reference time}} \right)}{\ln(f_{n+1}) - \ln(f_{n-1})} \quad (I)$$

ADIS enables examination of changes in the characteristic frequency (time constants) for each of the processes that change due to a change in operation conditions or to degradation.

3 Results

3.1 Durability of the Ni-ScYSZ based Solid Oxide Electrolysis Cell

After testing the initial performance of the reference cell, durability in electrolysis mode was examined at 800 °C with 51.4 % H₂O, 5.1 % H₂, 1.6 % CO, 15.8 % CO₂, and 26.1 % N₂ supplied to the Ni-ScYSZ electrode, oxygen supplied to the LSC-CGO electrode, and a current density of -0.25 A/cm². The evolution of cell voltage and corresponding in-plane with time for the test is shown in Figure 1.

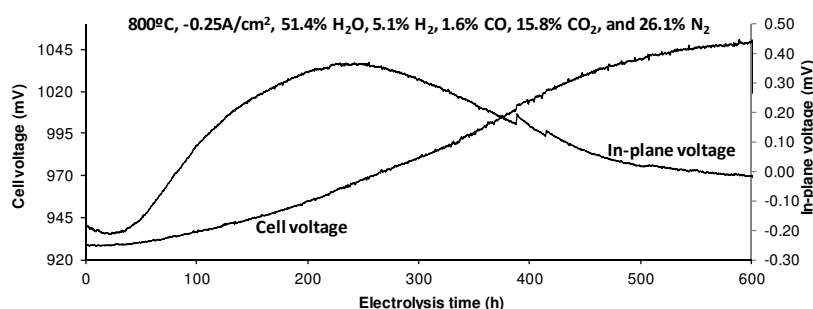


Figure 1. Cell voltage and corresponding in-plane voltage at the Ni-ScYSZ electrode measured for the Ni-ScYSZ based cell during electrolysis at -0.25 A.cm⁻² at 800 °C with 51.4 % H₂O, 5.1 % H₂, 1.6 % CO, 15.8 % CO₂, and 26.1 % N₂ supplied to the Ni-ScYSZ electrode, oxygen supplied to the LSC-CGO electrode.

From the cell voltage measured during electrolysis (Figure 1), it can be seen that the initial degradation was close to zero. The initial cell voltage during electrolysis was 930 mV and remained stable during the first 40 h of operation, hereafter is increased to 1035 mV after 475 h of operation corresponding to a degradation rate of ~220 mV / 1000 h. The in-plane voltage for the Ni-ScYSZ electrode (shown in Figure 1) indicates that the cell passivation/degradation may be a transient phenomenon as has previously been reported for passivation/activation during H₂O, CO₂ or co-electrolysis in SOECs⁷⁻⁹ and during sulphur poisoning of SOFCs¹⁰. The deposition of impurities on specific sites would create such a transient effect by a redistribution of the current as the cell resistance would be lower, where no impurities are deposited.

Based on the measured impedance, no change was observed in the ohmic (serial) resistance (R_s) whereas all cell degradation is caused by an increased polarisation resistance (R_p) as shown in Figure 2A. Analysis of the difference in impedance ADIS ($\frac{\Delta_t \partial Z'(f)}{\partial \ln(f)}$) during electrolysis was performed and is shown in Figure 2B. Due to initial difficulties in recording the impedance spectra, the spectrum recorded after 97 hours of electrolysis operation is used as the reference time for the ADIS as shown

in Figure 2. The differences in impedance spectra recorded during electrolysis show an initial increase at $\sim 200 - 300$ Hz, which shifted to ~ 80 Hz with time (Figure 2B).

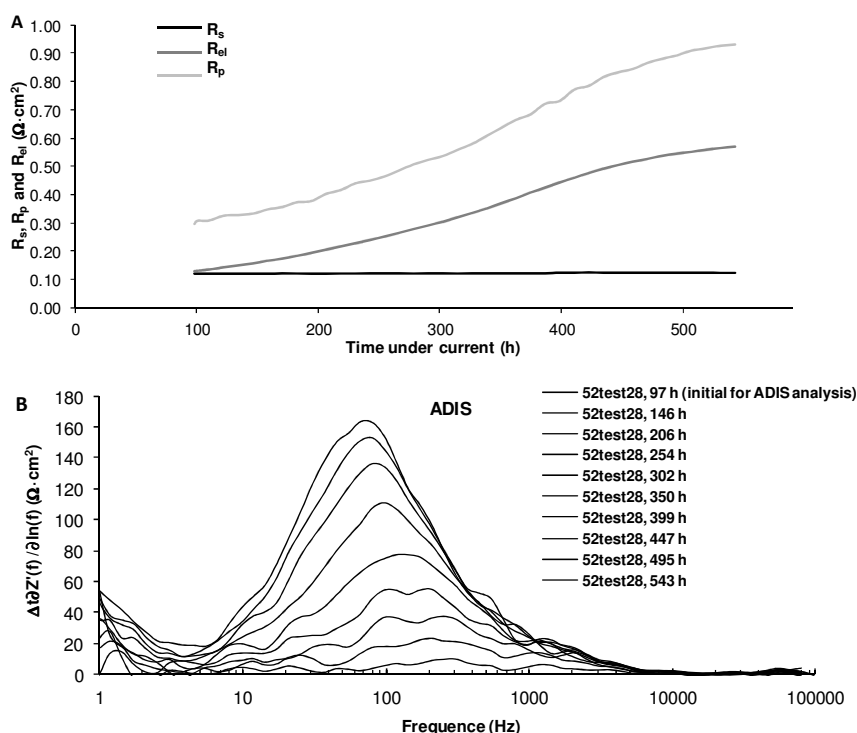


Figure 2. A) Ohmic resistance, R_s , electrochemical polarisation resistance, R_{el} , and total polarisation resistance, R_p . B) Analysis of the difference in impedance spectra (ADIS) during the passivation of the cell from 97 hours to 475 hours of electrolysis operation.

Based on the characteristic frequency, the passivation phenomena (80 – 200 Hz) may be assigned to a degradation of the Ni-ScYSZ electrode as previously suggested for Ni-YSZ based cells^{5,8,11-13} and may be due to a partial blockage of the TPB in the Ni-ScYSZ electrode caused by adsorption of impurities. That the degradation may be caused by the adsorption of impurities is supported by the evolution of cell voltage and in-plane voltage.

The cell voltage degradation is expected to level off, and indication of this can be seen in Figure 1 and Figure 2. Based on the analysis above, it seems like the cleaning did not remove all impurities. This was expected based on the method of cleaning. That not all impurities were removed creates a more realistic baseline measurement for the degradation of the SOCs operated on biogas.

3.2 Durability of the Solid Oxide Electrolysis Cells in the presence of sulphur

3.2.1 Cell voltage during electrolysis when introducing sulphur

After testing the initial performance of the cell (comparable to the reference test), durability in electrolysis mode was examined at 800 °C with 51.4 % H₂O, 5.1 % H₂, 1.6 % CO, 15.8 % CO₂, and 26.1 % N₂ (N₂ is sequentially substituted with CH₄) supplied to the Ni-ScYSZ electrode, oxygen supplied to the LSC-CGO electrode, and a current density of -0.25 A/cm². The total test was carried out for more than 850 hours (Figure 3), of which only the two periods with sulphur additions to the gas (first 265 hours, and the period from 535 to 795 hours) are described in this report.

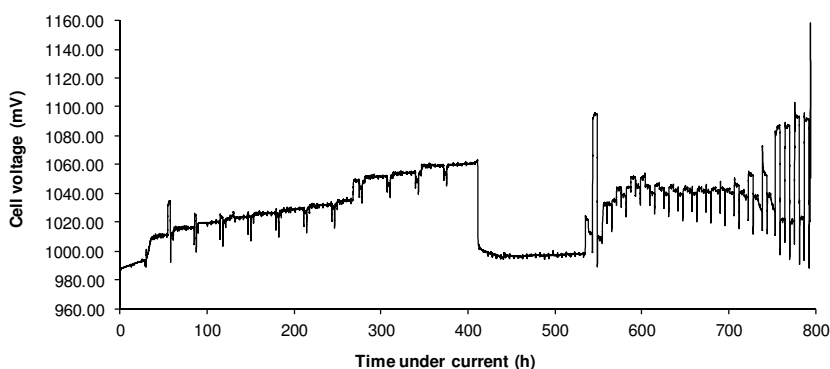


Figure 3. Cell voltage during the entire test. The increase in cell voltage after operation for 270 hours is caused by an increased hydrogen flow (due to a wrong initial calibration of the mass flow controller). The cell was operated at -0.25 A cm⁻² and at 800 °C with 51.4 % H₂O, 5.1 % H₂, 1.6 % CO, 15.8 % CO₂, and 26.1 % N₂ or CH₄ supplied to the Ni-ScYSZ electrode, and oxygen supplied to the LSC-CGO electrode.

After a period of 30 hours 2 ppm sulphur was added to the inlet gas. After additional 25 hours the nitrogen was substituted with methane. After introducing methane for 2.5 hours methane was removed from the inlet gas and exchanged with nitrogen. This sequence of durability with and without methane was repeated while adding 4 ppm, 6 ppm, 8 ppm, 10 ppm, 12 ppm, and 14 ppm sulphur, see Table 1. Beside probes for measuring the cell voltage, pO₂ probes were placed in the inlet and outlet streams. The pO₂ probes were constructed of single ended zirconia tube (supplied with a constant flow of air on the inside and with the inlet or outlet gas stream to or from the cell on the outside of the tube). Two Pt wires (one on the inside and one on the outside of the tube) measure the potential difference of the gas stream versus air which correlates to the partial pressure of oxygen in the stream. The probes measure the potential difference at the same temperature as the cell (800

°C). The measured potential difference versus air is denoted “ $E(pO_2)$ versus air in the inlet/outlet stream from the cell” in the following.

Table 1. Operating conditions, measured cell voltage, ASR and passivation during the cell test with the sulphur addition experiments. The numbers correspond to the numbers in **Error! Reference source not found.** (history plot of the cell voltage and $E(pO_2)$ versus air in the outlet stream from the cell at 800 °C vs. time). The measured cell voltage corresponds **Error! Reference source not found.** as well. ASRs are calculated from the measured cell voltage and applied current.

Time (h)	Gas composition to the Ni-ScYSZ electrode	Cell voltage (mV)	ASR _{cell voltage} ($\Omega \cdot \text{cm}^2$)	ASR _{Impedance spectroscopy} ($\Omega \cdot \text{cm}^2$)
0 – 30	H ₂ O/H ₂ /CO/CO ₂ /N ₂ 51.4/5.1/1.6/15.8/26	V _{1, start} = 987 V _{1, end} = 994	ASR _{1, start} = 0.38 ASR _{1, end} = 0.41	ASR _{1, start} = 0.38 ASR _{1, end} = 0.41
30 – 55	H ₂ O/H ₂ /CO/CO ₂ /N ₂ 51.4/5.1/1.6/15.8/26 + 2 ppm H ₂ S	V _{1, start} = 994 V _{1, end} = 1011	ASR _{1, start} = 0.41 ASR _{1, end} = 0.48	ASR _{1, start} = 0.41 ASR _{1, end} = 0.48
55 – 57.5	H ₂ O/H ₂ /CO/CO ₂ / CH₄ 51.4/5.1/1.6/15.8/26 + 2 ppm H ₂ S	V _{1, start} = 1032 V _{1, end} = 1035	ASR _{1, start} = 0.56 ASR _{1, end} = 0.57	ASR _{1, start} = 0.56 ASR _{1, end} = 0.57
57.5 – 60	H ₂ O/H ₂ /CO/CO ₂ / N₂ 51.4/5.1/1.6/15.8/26 + 2 ppm H ₂ S	V _{1, start} = 1010 V _{1, end} = 1011	ASR _{1, start} = 0.47 ASR _{1, end} = 0.48	ASR _{1, start} = 0.47 ASR _{1, end} = 0.48
60 – 85	H ₂ O/H ₂ /CO/CO ₂ /N ₂ 51.4/5.1/1.6/15.8/26 + 4 ppm H ₂ S	V _{1, start} = 1013 V _{1, end} = 1016	ASR _{1, start} = 0.49 ASR _{1, end} = 0.50	ASR _{1, start} = 0.49 ASR _{1, end} = 0.50
85 – 87.5	H ₂ O/H ₂ /CO/CO ₂ / CH₄ 51.4/5.1/1.6/15.8/26 + 4 ppm H ₂ S	V _{1, start} = 1026 V _{1, end} = 1025	ASR _{1, start} = 0.54 ASR _{1, end} = 0.54	ASR _{1, start} = 0.54 ASR _{1, end} = 0.54
87.5 – 90	H ₂ O/H ₂ /CO/CO ₂ / N₂ 51.4/5.1/1.6/15.8/26 + 4 ppm H ₂ S	V _{1, start} = 1014 V _{1, end} = 1017	ASR _{1, start} = 0.49 ASR _{1, end} = 0.50	ASR _{1, start} = 0.49 ASR _{1, end} = 0.50
90 – 115	H ₂ O/H ₂ /CO/CO ₂ / N₂ 51.4/5.1/1.6/15.8/26 + 6 ppm H ₂ S	V _{1, start} = 1019 V _{1, end} = 1020	ASR _{1, start} = 0.51 ASR _{1, end} = 0.52	ASR _{1, start} = 0.51 ASR _{1, end} = 0.52
115 – 117.5	H ₂ O/H ₂ /CO/CO ₂ / CH₄ 51.4/5.1/1.6/15.8/26 + 6ppm H ₂ S	V _{1, start} = 1023 V _{1, end} = 1023	ASR _{1, start} = 0.53 ASR _{1, end} = 0.53	ASR _{1, start} = 0.53 ASR _{1, end} = 0.53

117.5 122	-	H ₂ O/H ₂ /CO/CO ₂ / N ₂ 51.4/5.1/1.6/15.8/26 + 6 ppm H ₂ S	V _{1, start} = 1020 V _{1, end} = 1021	ASR _{1, start} = 0.52 ASR _{1, end} = 0.52	ASR _{1, start} = 0.52 ASR _{1, end} = 0.52
122 147.5	-	H ₂ O/H ₂ /CO/CO ₂ / N ₂ 51.4/5.1/1.6/15.8/26 + 8 ppm H ₂ S	V _{1, start} = 1022 V _{1, end} = 1023	ASR _{1, start} = 0.52 ASR _{1, end} = 0.53	ASR _{1, start} = 0.52 ASR _{1, end} = 0.53
147.5 150.5	-	H ₂ O/H ₂ /CO/CO ₂ / CH₄ 51.4/5.1/1.6/15.8/26 + 8 ppm H ₂ S	V _{1, start} = 1024 V _{1, end} = 1024	ASR _{1, start} = 0.53 ASR _{1, end} = 0.53	ASR _{1, start} = 0.53 ASR _{1, end} = 0.53
150.5 154	-	H ₂ O/H ₂ /CO/CO ₂ / N ₂ 51.4/5.1/1.6/15.8/26 + 8 ppm H ₂ S	V _{1, start} = 1023 V _{1, end} = 1024	ASR _{1, start} = 0.53 ASR _{1, end} = 0.53	ASR _{1, start} = 0.53 ASR _{1, end} = 0.53
154 – 180		H ₂ O/H ₂ /CO/CO ₂ / N ₂ 51.4/5.1/1.6/15.8/26 + 10 ppm H ₂ S	V _{1, start} = 1025 V _{1, end} = 1027	ASR _{1, start} = 0.54 ASR _{1, end} = 0.54	ASR _{1, start} = 0.54 ASR _{1, end} = 0.54
180 181.5	-	H ₂ O/H ₂ /CO/CO ₂ / CH₄ 51.4/5.1/1.6/15.8/26 + 10 ppm H ₂ S	V _{1, start} = 1025 V _{1, end} = 1025	ASR _{1, start} = 0.54 ASR _{1, end} = 0.54	ASR _{1, start} = 0.54 ASR _{1, end} = 0.54
181.5 186	-	H ₂ O/H ₂ /CO/CO ₂ / N ₂ 51.4/5.1/1.6/15.8/26 + 10 ppm H ₂ S	V _{1, start} = 1026 V _{1, end} = 1027	ASR _{1, start} = 0.54 ASR _{1, end} = 0.54	ASR _{1, start} = 0.54 ASR _{1, end} = 0.54
186 – 212		H ₂ O/H ₂ /CO/CO ₂ / N ₂ 51.4/5.1/1.6/15.8/26 + 12 ppm H ₂ S	V _{1, start} = 1028 V _{1, end} = 1029	ASR _{1, start} = 0.55 ASR _{1, end} = 0.55	ASR _{1, start} = 0.55 ASR _{1, end} = 0.55
212 212.5	-	H ₂ O/H ₂ /CO/CO ₂ / CH₄ 51.4/5.1/1.6/15.8/26 + 12 ppm H ₂ S	V _{1, start} = 1027 V _{1, end} = 1027	ASR _{1, start} = 0.54 ASR _{1, end} = 0.54	ASR _{1, start} = 0.54 ASR _{1, end} = 0.54
212.5 218	-	H ₂ O/H ₂ /CO/CO ₂ / N ₂ 51.4/5.1/1.6/15.8/26 + 12 ppm H ₂ S	V _{1, start} = 1028 V _{1, end} = 1030	ASR _{1, start} = 0.55 ASR _{1, end} = 0.56	ASR _{1, start} = 0.55 ASR _{1, end} = 0.56
218 – 244		H ₂ O/H ₂ /CO/CO ₂ / N ₂ 51.4/5.1/1.6/15.8/26 + 14 ppm H ₂ S	V _{1, start} = 1031 V _{1, end} = 1032	ASR _{1, start} = 0.56 ASR _{1, end} = 0.56	ASR _{1, start} = 0.56 ASR _{1, end} = 0.56
244 245.5	-	H ₂ O/H ₂ /CO/CO ₂ / CH₄ 51.4/5.1/1.6/15.8/26 + 14 ppm H ₂ S	V _{1, start} = 1030 V _{1, end} = 1030	ASR _{1, start} = 0.56 ASR _{1, end} = 0.56	ASR _{1, start} = 0.56 ASR _{1, end} = 0.56

245.5 250.5	–	H ₂ O/H ₂ /CO/CO ₂ / N ₂ 51.4/5.1/1.6/15.8/26 + 14 ppm H ₂ S	V _{1, start} = 1032 V _{1, end} = 1033	ASR _{1, start} = 0.56 ASR _{1, end} = 0.57	ASR _{1, start} = 0.56 ASR _{1, end} = 0.57
250.5 265	–	H ₂ O/H ₂ /CO/CO ₂ / N ₂ 51.4/5.1/1.6/15.8/26 + 16 ppm H ₂ S	V _{1, start} = 1034 V _{1, end} = 1035	ASR _{1, start} = 0.57 ASR _{1, end} = 0.58	ASR _{1, start} = 0.57 ASR _{1, end} = 0.58

The evolution of cell voltage and $E(pO_2)$ versus air in the outlet stream from the cell with time for the test is shown in Fig. 4. From the cell voltage measured during electrolysis (Fig. 4), it can be seen that the initial degradation was increased compared to the reference test. The increase in initial degradation may be a consequence of the attempts to clean the gases for the reference test (which may have decreased the initial degradation) or due to a slightly higher initial performance for the reference test. The initial cell voltage during electrolysis was 987 mV and increased to 994 mV during the first 30 h of operation. Hereafter 2 ppm H₂S was introduced which caused the cell voltage to increase to 1008 mV after 5 hours of sulphur addition; hereafter the cell voltage increased only little (increased to 1010 mV after 55 h of operation). The in-plane voltage for the Ni-ScYSZ electrode (not shown) show the characteristic S-shape change which is observed for poisoning by impurities as shown for the reference test and has previously been reported for passivation/activation during H₂O, CO₂ or co-electrolysis in SOECs⁷⁻⁹ and during sulphur poisoning of SOFCs¹⁰. The adsorption of impurities on active sites moves gradually in the flow direction of the steam and CO₂ as a kind of front and creates such a transient effect by an uneven redistribution of the current density.

When nitrogen with 2 ppm H₂S was replaced with CH₄ 2 ppm H₂S the cell voltage immediately increased to 1032 mV (Fig. 4A). This indicates that some methane was converted to carbon monoxide and hydrogen (steam and CO₂ reforming) and by this decreases the p_{H₂O} and p_{CO₂}. The lower concentrations of reactants cause a high overvoltage in order to maintain the constant current density. This is further supported by the decrease in $E(pO_2)$ versus air in the outlet stream from the cell, which would otherwise remain stable (Figure 4B). After the introduction of methane for 2.5 hours, the methane was again replaced with nitrogen and the cell voltage decreased to its original value (1010 mV, Table 1 and Fig. 4A). Hereafter 4 ppm H₂S was introduced to the cell which again caused the cell voltage to increase. The increase in cell voltage was not as drastic as when introducing sulphur to the “fresh” cell, and increased by only 3 mV. When flowing 4 ppm H₂S in nitrogen was again replaced with 4 ppm H₂S in CH₄ the cell voltage immediately increase to 1026 mV (Fig. 4A) indicating that some methane and steam (and CO₂) was converted to carbon monoxide and hydrogen.

Again this is supported by the decrease in $E(pO_2)$ versus air in the outlet stream from the cell. It has to be noticed that the increase in cell voltage and decrease in $E(pO_2)$ versus air in the outlet stream from the cell when introducing methane while flowing 4 ppm sulphur ($\Delta_{\text{cell}} \text{ voltage} = 10 \text{ mV}$, and $\Delta E(pO_2)$ of 36 mV) is smaller than when introducing methane while flowing only 2 ppm sulphur ($\Delta_{\text{cell}} \text{ voltage} = 21 \text{ mV}$, and $\Delta E(pO_2)$ of 52 mV). During the subsequent sequences where 6, 8, 10, and 12 ppm sulphur was added with CH_4 . The $\Delta_{\text{cell}} \text{ voltage}$ decreased only few mV and the $\Delta E(pO_2)$ decreased with $16 \text{ mV} \pm 1 \text{ mV}$ and remained constant when introducing 10 ppm or higher concentrations of sulphur to the cell. The sensor measuring $E(pO_2)$ versus air in the outlet stream is made of Pt, and it seems as if the CH_4 reforming on Pt is poisoned less by sulphur than Ni. The Pt electrode of the oxygen monitor senses a reducing effect (increasing $(H_2 + CO/H_2O + CO_2)$ ratio) of the CH_4 even at 14 ppm, whereas the electrolysis on the Ni seems unaffected by any possible reforming of CH_4 already at 8 ppm S.

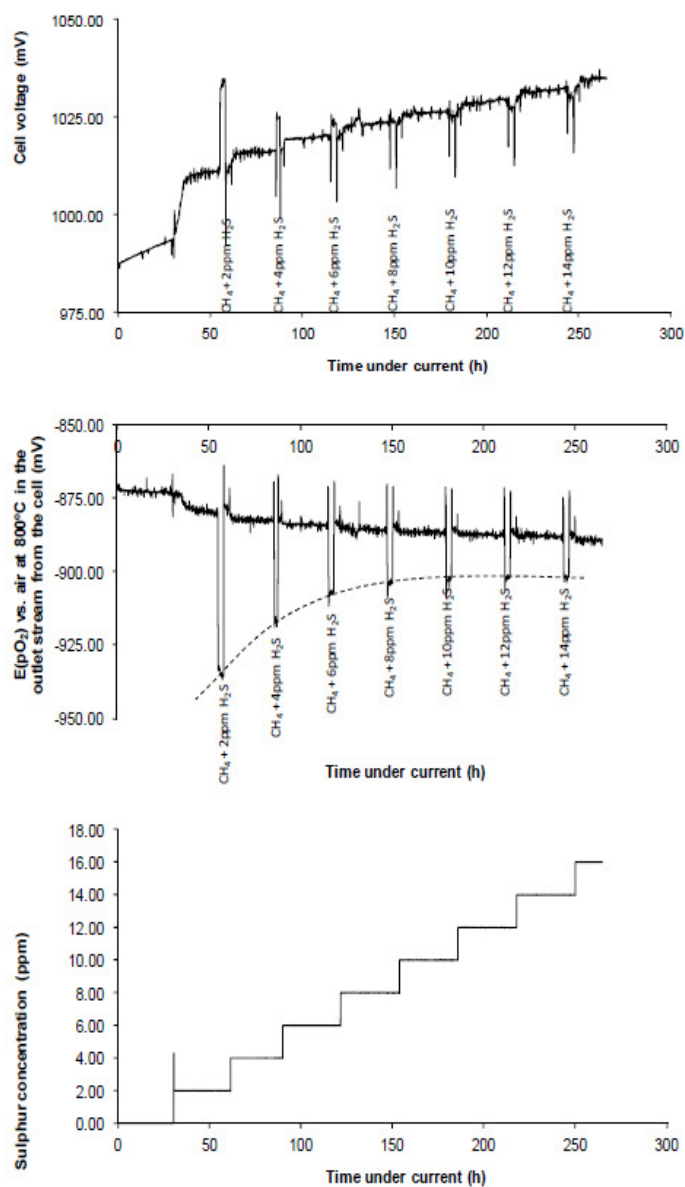


Figure 4. A: Cell voltage, see table 1 for details regarding the changes in gas composition, and B: $E(pO_2)$ versus air in the outlet stream from the cell at 800 °C measured during electrolysis at -0.25 A cm^{-2} at 800 °C with 51.4 % H_2O , 5.1 % H_2 , 1.6 % CO , 15.8 % CO_2 , and 26.1 % N_2 or CH_4 supplied to the Ni-ScYSZ electrode, and oxygen supplied to the LSC-CGO electrode.

3.2.2 Electrochemical impedance spectroscopy during electrolysis applying gasses as received

To investigate the degradation, impedance spectra were recorded during electrolysis. Figure 4 show the evolution in the serial resistance and total polarisation resistance, R_p . Further, the electrochemical polarisation resistance, R_{el} , is shown in the figure.

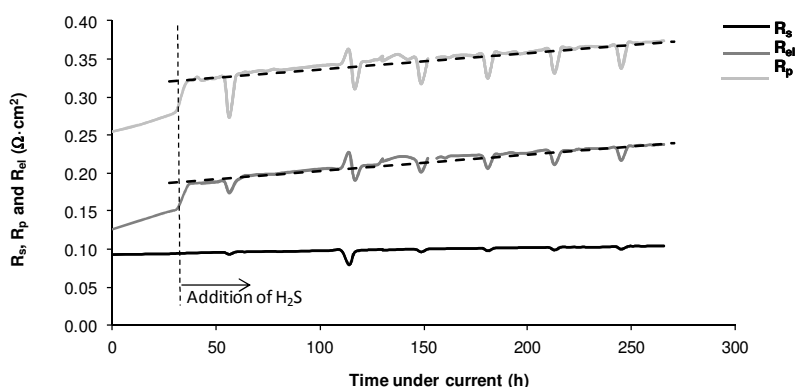


Figure 4. Evolution in the serial resistance, polarisation resistance and the electrochemical polarisation resistance during the first 265 hours of test.

Examination of the evolution in impedance during the degradation of the SOECs, reveals that the polarisation resistance increases significant and fast when introducing 2 ppm sulphur. Beside the increase when introducing 2ppm sulphur, no sudden increase in polarisation was observed when increasing the sulphur concentration. On the other hand, when increasing the sulphur concentration, a graduate increase in polarisation resistance was observed. It should be notices that the rate of the increase in polarisation resistance is not higher than the initial rate of increase in polarisation resistance, which may indicate that sulphur adsorbs on the most active sites during the introduction of 2 ppm sulphur whereafter the slightly less active sites are operating almost unaffected. The evolution in impedance is further analysed by ADIS as shown in Figure 5. The ADIS analysis shows that the same passivation/degradation processes occur during the introduction of sulphur as when operating the reference test (Figure 2B) with an increase in resistance around 200 – 300 Hz. Since sulphur was introduced in this test, the increase may be assigned to a partial blockage of the TPB in the Ni-ScYSZ electrode caused by adsorption of sulphur and supports the hypothesis that the durability of the reference test was also influenced by the adsorption of gas impurities at the Ni-ScYSZ electrode. It looks as the degradation rates of the reference cell and of the sulphur experiment cell are similar, i.e. a “background” degradation rate, and that the degradation rate (apart from the first

fast step) due to the added sulphur is small. This might be taken as an indication that the background degradation rate is due to another (unknown) impurity than sulphur, but this should be investigated further.

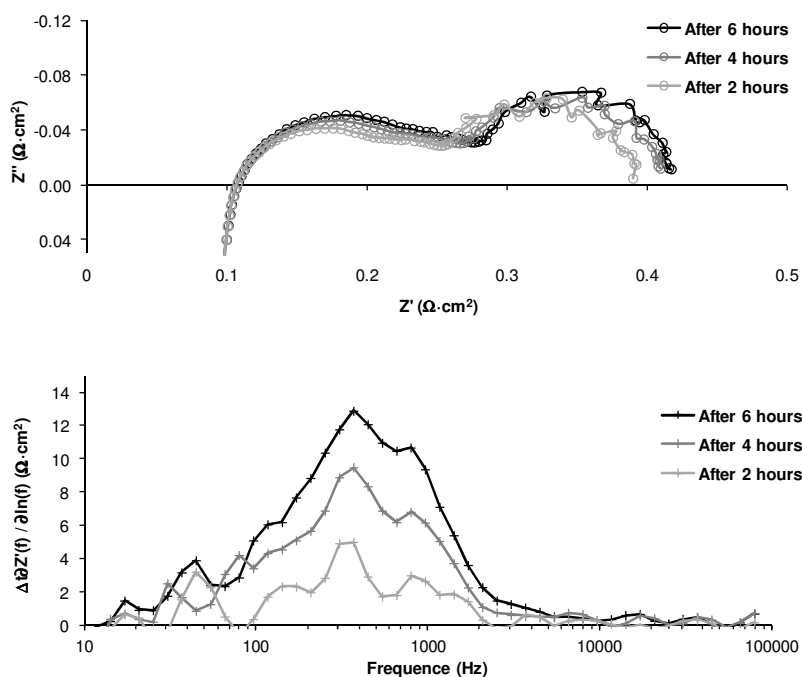


Figure 5. Top: Nyquist plot of impedance spectra recorded during introduction of sulphur. Bottom: Analysis of the difference in impedance spectra during introduction of H_2S .

The impedance contribution may be separated into the serial resistance and electrode polarisation resistance. The polarisation resistance may be further separated into the electrochemical polarisation and a contribution originating from the conversion of gases as a consequence of the change in EMF (the so-called gas conversion resistance). The conversion resistance is dependent on the gas composition, and is related to the reactant/product concentration (and current density). If no steam reforming occurs, the gas conversion impedance is theoretical identical in the mixture containing nitrogen ($\text{H}_2\text{O}/\text{H}_2/\text{CO}/\text{CO}_2/\text{N}_2$) and the mixture containing methane ($\text{H}_2\text{O}/\text{H}_2/\text{CO}/\text{CO}_2/\text{CH}_4$) since methane will pass through the cell without being converted (similar to nitrogen). On the other hand, if steam reforming occurs, methane will be converted and will in this case not be inert. The increase in reactants/products will thus lower the conversion resistance, when the gas composition is approaching a ratio of $(\text{H}_2 + \text{CO})/(\text{H}_2\text{O} + \text{CO}_2)$ of 1. The measured conversion resistance during the first 265 hours of test is shown in

Figure 6.

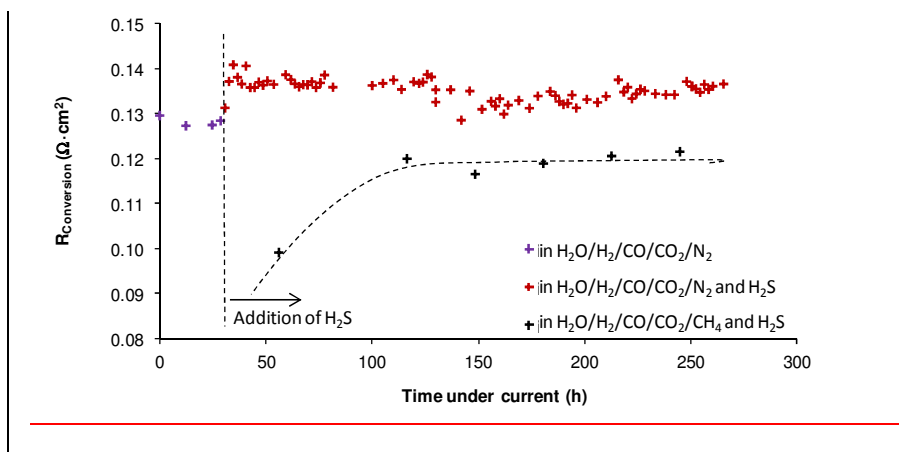


Figure 6. Evolution in the conversion resistance during the first 265 hours of test.

The decrease in conversion resistance when substituting nitrogen with methane might indicate that some methane is converted due to the steam reforming reaction. However, the size of the changes is close to the uncertainty on the conversion polarisation due to uncertainty of the flow control. This seems to occur throughout the test, although to a less extent when introducing 6 ppm or higher concentrations of sulphur to the cell. After the first sulphur experiment, the cell was operated without sulphur for 125 hours (after operation for 411 hours) to re-activate the cell (remove adsorbed sulphur according to $S_{(ads)} + H_{2(gas)} \rightarrow H_{2S(gas)}$). After the re-activation period a second sulphur experiment was carried out on the same cell. This sulphur experiment was similar to the first experiment, although the sulphur concentration was rapidly increased to 15 ppm (with a step of 5 ppm) whereafter the sulphur concentration was slowly decreased. First the sulphur concentration was decreased to 10 ppm, whereafter the concentration was decreased stepwise (with a step of 2 ppm) to 6ppm. The final decrease in sulphur concentration to zero ppm was carried out with a step of 1 ppm. After removing the sulphur from the inlet flow to the cell (after 748 hours of operation), four periods with methane was carried out, see Figure 7. The evolution of cell voltage and $E(pO_2)$ versus air in the outlet stream from the cell as well as the methane and sulphur concentration for the 2nd test is shown in Figure 7.

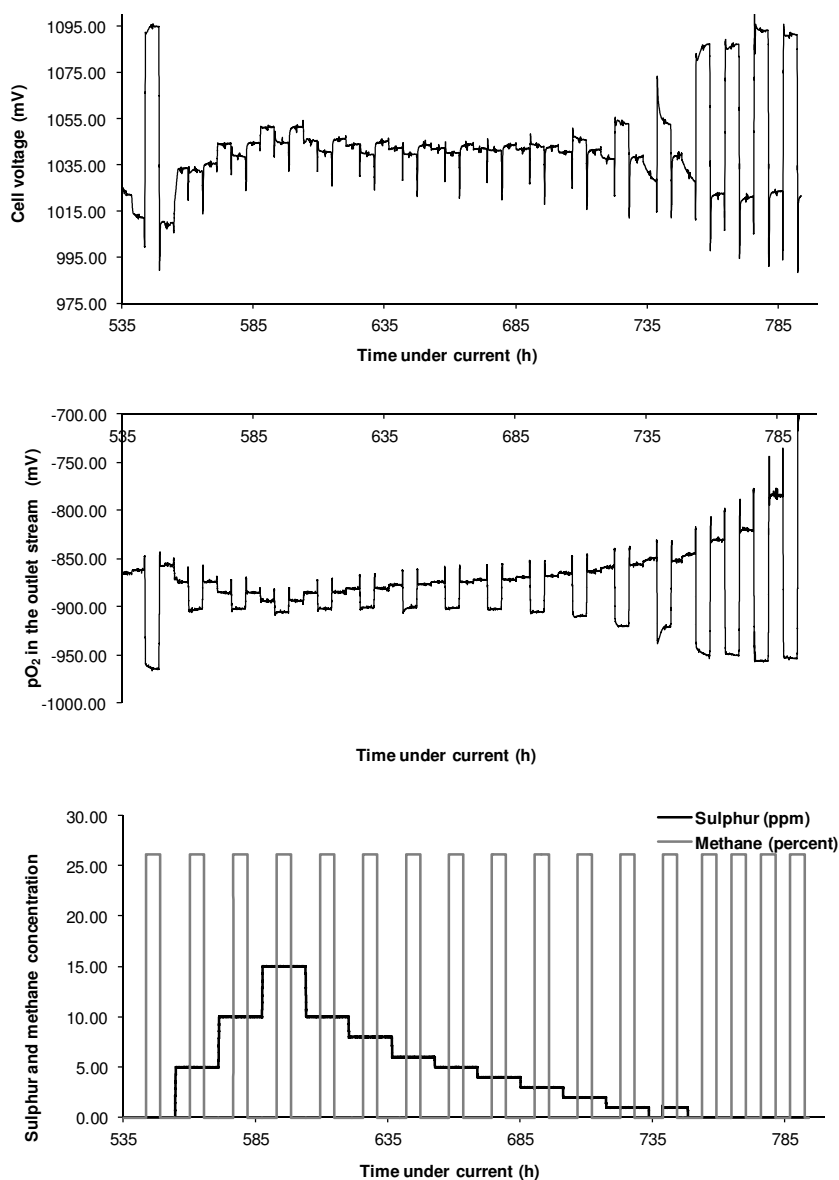


Figure 7. A: Cell voltage, B: $E(pO_2)$ versus air in the outlet stream from the cell, and C: Methane and sulphur concentration for the 2nd sulphur experiment. The cell was operated at -0.25 Acm^{-2} at $800 \text{ }^\circ\text{C}$ with 51.4 % H_2O , 5.1 % H_2 , 1.6 % CO , 15.8 % CO_2 , and 26.1 % N_2 or CH_4 supplied to the Ni-ScYSZ electrode, and oxygen supplied to the LSC-CGO electrode.

The cell voltage after recovery was $\sim 1010 - 1020 \text{ mV}$ whereas the initial cell voltage was around 990 mV (Figure 3, Figure 4, Figure 7 and Table 1, Table 2). This shows that the cell voltage did not completely recover after the period without sulphur.

After the initial operation, nitrogen was replaced with CH_4 which immediately caused the cell voltage to increase similar to the first experiment. This indicates that some methane was converted to carbon monoxide and hydrogen (steam reforming). This is further supported by the decrease in $E(\text{pO}_2)$ versus air in the outlet stream from the cell, which would otherwise remain stable. Hereafter sulphur was introduced and nitrogen was again replaced with CH_4 which once again caused the cell voltage to increase. These series was repeated while first increasing the sulphur concentration and thereafter decreasing the sulphur concentration once again.

The change in cell voltage ($\Delta_{\text{cell voltage}}$) and change in $E(\text{pO}_2)$ versus air in the outlet stream from the cell ($\Delta E(\text{pO}_2)$) when introducing methane decreased with increasing sulphur concentration up to around 2 – 4 ppm, whereas at higher sulphur concentrations $\Delta_{\text{cell voltage}}$ and $\Delta E(\text{pO}_2)$ changed only very little. This second experiment confirms the initial experiment and clearly shows that the presence of sulphur reduces the steam reforming activity, and decreased to a stable level at sulphur concentrations above 2 – 4 ppm.

3.3 Characterisation of metal supported cells

Since the nickel loading is much lower in the metal supported they are presumed to possess a better sulphur tolerance and it was therefore the attempt to apply a metal supported cell for the upgrading of the biogas. After reduction, the metal supported cell was characterised in $\text{H}_2\text{O} - \text{H}_2$ mixtures. Because of corrosion issues for these metal supported cells, the cell was operated at maximal 800 °C and with less oxidising conditions than the standard procedure. The procedure consists of AC and DC characterisation in the temperature range from 650 °C to 800 °C with various gas mixtures supplied to the cathode (4 % $\text{H}_2\text{O} - 96\% \text{H}_2$, 20 % $\text{H}_2\text{O} - 80\% \text{H}_2$, 50 % $\text{H}_2\text{O} - 50\% \text{H}_2$), and pure oxygen or air supplied to the anode. Further, additional AC and DC characterisation was performed in simulated biogas supplied to the cathode, only at 650 °C. The initial performance of the cell at 800 °C is shown in Figure 8. From Figure 8 it can be seen that the initial performance of the metal supported is higher than the initial performance for the Ni-ScYSZ based SOC as used for the reference test as well as the sulphur experiment. It was intended to operate this cell similar to the Ni-ScYSZ based SOC. Unfortunately the cell voltage increased drastically after ca. one day of test with almost stable voltage. The cell has not yet been examined post mortem and the specific reason for the fast degradation is therefore not known with certainty.

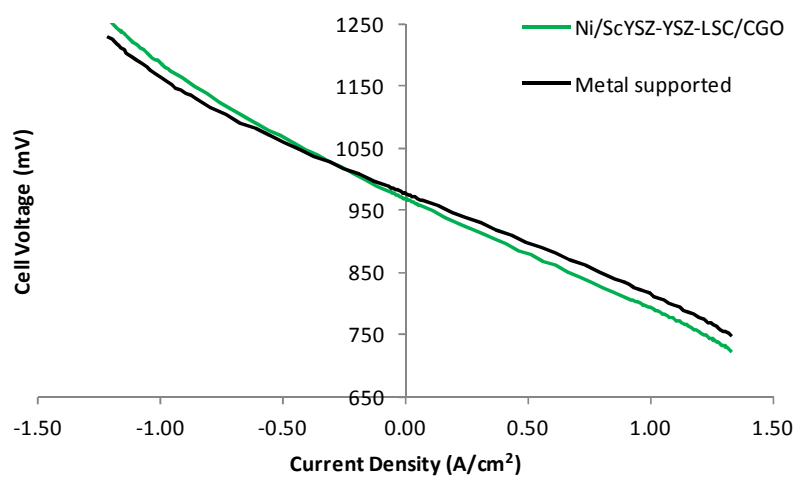


Figure 8. Initial DC characterisation for the metal supported cell and the Ni-ScYSZ based cell in 50 % H₂O + 50 % H₂ at 800°C.

Table 2. Operating conditions, measured cell voltage, ASR and passivation during the durability test for cell-3. The measured cell voltage corresponds to Figure 7. ASRs are calculated from the measured cell voltage and applied current.

Time (h)	Gas composition to the Ni-ScYSZ electrode	Cell voltage (mV)	ASR _{cell voltage} ($\Omega \cdot \text{cm}^2$)
535 – 543	H ₂ O/H ₂ /CO/CO ₂ /N ₂ 51.4/5.1/1.6/15.8/26	V _{start} = 1024 V _{end} = 1011	ASR _{start} = 0.53 ASR _{end} = 0.48
543 - 549	H ₂ O/H ₂ /CO/CO ₂ / CH₄ 51.4/5.1/1.6/15.8/26	V _{start} = 1092 V _{end} = 1095	ASR _{start} = 0.80 ASR _{end} = 0.82
549 – 554	H ₂ O/H ₂ /CO/CO ₂ /N ₂ 51.4/5.1/1.6/15.8/26	V _{start} = 1009 V _{end} = 1010	ASR _{start} = 0.47 ASR _{end} = 0.48
554 – 560	H ₂ O/H ₂ /CO/CO ₂ /N ₂ 51.4/5.1/1.6/15.8/26 + 5 ppm H ₂ S	V _{start} = 1019 V _{end} = 1033	ASR _{start} = 0.51 ASR _{end} = 0.57
560 – 565	H ₂ O/H ₂ /CO/CO ₂ / CH₄ 51.4/5.1/1.6/15.8/26 + 5 ppm H ₂ S	V _{start} = 1031 V _{end} = 1032	ASR _{start} = 0.56 ASR _{end} = 0.56
565 - 570	H ₂ O/H ₂ /CO/CO ₂ / N₂ 51.4/5.1/1.6/15.8/26 + 5 ppm H ₂ S	V _{start} = 1034 V _{end} = 1035	ASR _{start} = 0.57 ASR _{end} = 0.58
570 - 576	H ₂ O/H ₂ /CO/CO ₂ /N ₂ 51.4/5.1/1.6/15.8/26 + 10 ppm H ₂ S	V _{start} = 1043 V _{end} = 1044	ASR _{start} = 0.61 ASR _{end} = 0.61
576 - 581	H ₂ O/H ₂ /CO/CO ₂ / CH₄ 51.4/5.1/1.6/15.8/26 + 10 ppm H ₂ S	V _{start} = 1038 V _{end} = 1038	ASR _{start} = 0.59 ASR _{end} = 0.59
581 – 587	H ₂ O/H ₂ /CO/CO ₂ / N₂ 51.4/5.1/1.6/15.8/26 + 10 ppm H ₂ S	V _{start} = 1044 V _{end} = 1044	ASR _{start} = 0.61 ASR _{end} = 0.61
587 - 593	H ₂ O/H ₂ /CO/CO ₂ / N₂ 51.4/5.1/1.6/15.8/26 + 15 ppm H ₂ S	V _{start} = 1051 V _{end} = 1051	ASR _{start} = 0.64 ASR _{end} = 0.64

593 – 598	H ₂ O/H ₂ /CO/CO ₂ / CH₄ 51.4/5.1/1.6/15.8/26 + 15 ppm H ₂ S	V _{start} = 1044 V _{end} = 1044	ASR _{start} = 0.61 ASR _{end} = 0.61
598 – 604	H ₂ O/H ₂ /CO/CO ₂ / N ₂ 51.4/5.1/1.6/15.8/26 + 15 ppm H ₂ S	V _{start} = 1051 V _{end} = 1051	ASR _{start} = 0.64 ASR _{end} = 0.64
604 – 610	H ₂ O/H ₂ /CO/CO ₂ / N ₂ 51.4/5.1/1.6/15.8/26 + 10 ppm H ₂ S	V _{start} = 1045 V _{end} = 1045	ASR _{start} = 0.62 ASR _{end} = 0.62
610 – 615	H ₂ O/H ₂ /CO/CO ₂ / CH₄ 51.4/5.1/1.6/15.8/26 + 10 ppm H ₂ S	V _{start} = 1041 V _{end} = 1041	ASR _{start} = 0.60 ASR _{end} = 0.60
615 – 620	H ₂ O/H ₂ /CO/CO ₂ / N ₂ 51.4/5.1/1.6/15.8/26 + 10 ppm H ₂ S	V _{start} = 1046 V _{end} = 1046	ASR _{start} = 0.62 ASR _{end} = 0.62
620 – 625	H ₂ O/H ₂ /CO/CO ₂ / N ₂ 51.4/5.1/1.6/15.8/26 + 8 ppm H ₂ S	V _{start} = 1043 V _{end} = 1043	ASR _{start} = 0.61 ASR _{end} = 0.61
625 – 630	H ₂ O/H ₂ /CO/CO ₂ / CH₄ 51.4/5.1/1.6/15.8/26 + 8 ppm H ₂ S	V _{start} = 1039 V _{end} = 1039	ASR _{start} = 0.59 ASR _{end} = 0.59
630 – 636	H ₂ O/H ₂ /CO/CO ₂ / N ₂ 51.4/5.1/1.6/15.8/26 + 8 ppm H ₂ S	V _{start} = 1044 V _{end} = 1045	ASR _{start} = 0.61 ASR _{end} = 0.62
636 – 642	H ₂ O/H ₂ /CO/CO ₂ / N ₂ 51.4/5.1/1.6/15.8/26 + 6 ppm H ₂ S	V _{start} = 1042 V _{end} = 1042	ASR _{start} = 0.60 ASR _{end} = 0.60
642 – 647	H ₂ O/H ₂ /CO/CO ₂ / CH₄ 51.4/5.1/1.6/15.8/26 + 6 ppm H ₂ S	V _{start} = 1039 V _{end} = 1039	ASR _{start} = 0.59 ASR _{end} = 0.59
647 – 652	H ₂ O/H ₂ /CO/CO ₂ / N ₂ 51.4/5.1/1.6/15.8/26 + 6 ppm H ₂ S	V _{start} = 1042 V _{end} = 1043	ASR _{start} = 0.60 ASR _{end} = 0.61
652 – 658	H ₂ O/H ₂ /CO/CO ₂ / N ₂ 51.4/5.1/1.6/15.8/26 + 5 ppm H ₂ S	V _{start} = 1042 V _{end} = 1042	ASR _{start} = 0.60 ASR _{end} = 0.60

658 – 663	H ₂ O/H ₂ /CO/CO ₂ / CH₄ 51.4/5.1/1.6/15.8/26 + 5 ppm H ₂ S	V _{start} = 1040 V _{end} = 1040	ASR _{start} = 0.60 ASR _{end} = 0.60
663 – 669	H ₂ O/H ₂ /CO/CO ₂ / N ₂ 51.4/5.1/1.6/15.8/26 + 5 ppm H ₂ S	V _{start} = 1042 V _{end} = 1043	ASR _{start} = 0.60 ASR _{end} = 0.61
669 – 674	H ₂ O/H ₂ /CO/CO ₂ / N ₂ 51.4/5.1/1.6/15.8/26 + 4 ppm H ₂ S	V _{start} = 1042 V _{end} = 1042	ASR _{start} = 0.60 ASR _{end} = 0.60
674 – 679	H ₂ O/H ₂ /CO/CO ₂ / CH₄ 51.4/5.1/1.6/15.8/26 + 4 ppm H ₂ S	V _{start} = 1041 V _{end} = 1041	ASR _{start} = 0.60 ASR _{end} = 0.60
679 – 685	H ₂ O/H ₂ /CO/CO ₂ / N ₂ 51.4/5.1/1.6/15.8/26 + 4 ppm H ₂ S	V _{start} = 1043 V _{end} = 1043	ASR _{start} = 0.61 ASR _{end} = 0.61
685 – 690	H ₂ O/H ₂ /CO/CO ₂ / N ₂ 51.4/5.1/1.6/15.8/26 + 3 ppm H ₂ S	V _{start} = 1041 V _{end} = 1042	ASR _{start} = 0.60 ASR _{end} = 0.60
690 – 695	H ₂ O/H ₂ /CO/CO ₂ / CH₄ 51.4/5.1/1.6/15.8/26 + 3 ppm H ₂ S	V _{start} = 1044 V _{end} = 1043	ASR _{start} = 0.61 ASR _{end} = 0.61
695 – 701	H ₂ O/H ₂ /CO/CO ₂ / N ₂ 51.4/5.1/1.6/15.8/26 + 3 ppm H ₂ S	V _{start} = 1042 V _{end} = 1043	ASR _{start} = 0.60 ASR _{end} = 0.61
701 – 706	H ₂ O/H ₂ /CO/CO ₂ / N ₂ 51.4/5.1/1.6/15.8/26 + 2 ppm H ₂ S	V _{start} = 1040 V _{end} = 1040	ASR _{start} = 0.60 ASR _{end} = 0.60
706 – 712	H ₂ O/H ₂ /CO/CO ₂ / CH₄ 51.4/5.1/1.6/15.8/26 + 2 ppm H ₂ S	V _{start} = 1047 V _{end} = 1046	ASR _{start} = 0.62 ASR _{end} = 0.62
712 – 717	H ₂ O/H ₂ /CO/CO ₂ / N ₂ 51.4/5.1/1.6/15.8/26 + 2 ppm H ₂ S	V _{start} = 1040 V _{end} = 1041	ASR _{start} = 0.60 ASR _{end} = 0.60
717 – 722	H ₂ O/H ₂ /CO/CO ₂ / N ₂ 51.4/5.1/1.6/15.8/26 + 1 ppm H ₂ S	V _{start} = 1038 V _{end} = 1037	ASR _{start} = 0.59 ASR _{end} = 0.58

722 – 728	H ₂ O/H ₂ /CO/CO ₂ / CH₄ 51.4/5.1/1.6/15.8/26 + 1 ppm H ₂ S	V _{start} = 1053 V _{end} = 1053	ASR _{start} = 0.65 ASR _{end} = 0.65
728 – 734	H ₂ O/H ₂ /CO/CO ₂ / N ₂ 51.4/5.1/1.6/15.8/26 + 1 ppm H ₂ S	V _{start} = 1037 V _{end} = 1038	ASR _{start} = 0.58 ASR _{end} = 0.59
734 – 739	H ₂ O/H ₂ /CO/CO ₂ / N ₂ 51.4/5.1/1.6/15.8/26 + 0 ppm H ₂ S	V _{start} = 1035 V _{end} = 1027	ASR _{start} = 0.58 ASR _{end} = 0.54
739 – 744	H ₂ O/H ₂ /CO/CO ₂ / CH₄ 51.4/5.1/1.6/15.8/26 + 1 ppm H ₂ S	V _{start} = 1070 V _{end} = 1052	ASR _{start} = 0.72 ASR _{end} = 0.64
744 – 748	H ₂ O/H ₂ /CO/CO ₂ / N ₂ 51.4/5.1/1.6/15.8/26 + 1 ppm H ₂ S	V _{start} = 1038 V _{end} = 1039	ASR _{start} = 0.59 ASR _{end} = 0.59
748 – 753	H ₂ O/H ₂ /CO/CO ₂ / N ₂ 51.4/5.1/1.6/15.8/26 + 0 ppm H ₂ S	V _{start} = 1037 V _{end} = 1027	ASR _{start} = 0.58 ASR _{end} = 0.54
753 – 759	H ₂ O/H ₂ /CO/CO ₂ / CH₄ 51.4/5.1/1.6/15.8/26 + 0 ppm H ₂ S	V _{start} = 1080 V _{end} = 1087	ASR _{start} = 0.76 ASR _{end} = 0.78
759 – 764	H ₂ O/H ₂ /CO/CO ₂ / N ₂ 51.4/5.1/1.6/15.8/26 + 0 ppm H ₂ S	V _{start} = 1022 V _{end} = 1022	ASR _{start} = 0.52 ASR _{end} = 0.52
764 – 770	H ₂ O/H ₂ /CO/CO ₂ / CH₄ 51.4/5.1/1.6/15.8/26 + 0 ppm H ₂ S	V _{start} = 1087 V _{end} = 1087	ASR _{start} = 0.78 ASR _{end} = 0.78
770 – 775	H ₂ O/H ₂ /CO/CO ₂ / N ₂ 51.4/5.1/1.6/15.8/26 + 0 ppm H ₂ S	V _{start} = 1019 V _{end} = 1020	ASR _{start} = 0.51 ASR _{end} = 0.52
775 – 781	H ₂ O/H ₂ /CO/CO ₂ / CH₄ 51.4/5.1/1.6/15.8/26 + 0 ppm H ₂ S	V _{start} = 1094 V _{end} = 1094	ASR _{start} = 0.81 ASR _{end} = 0.81
781 – 787	H ₂ O/H ₂ /CO/CO ₂ / N ₂ 51.4/5.1/1.6/15.8/26 + 0 ppm H ₂ S	V _{start} = 1022 V _{end} = 1023	ASR _{start} = 0.52 ASR _{end} = 0.53

787 – 792	H ₂ O/H ₂ /CO/CO ₂ / CH₄ 51.4/5.1/1.6/15.8/26 + 0 ppm H ₂ S	V _{start} = 1092 V _{end} = 1091	ASR _{start} = 0.80 ASR _{end} = 0.80
792 –	H ₂ O/H ₂ /CO/CO ₂ / N ₂ 51.4/5.1/1.6/15.8/26 + 0 ppm H ₂ S	V _{start} = 1021	ASR _{start} = 0.52

4 Conclusions and Outlook

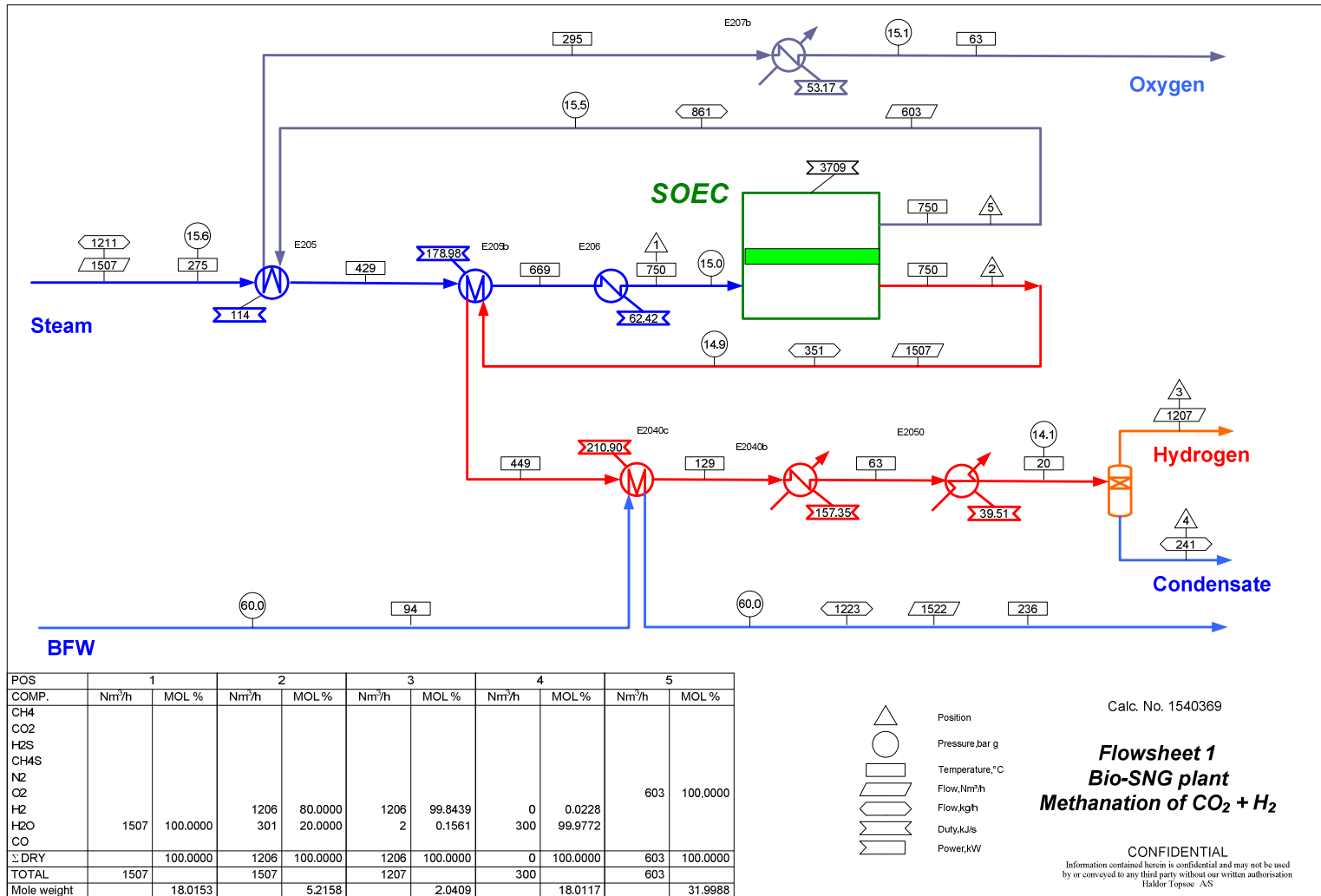
The electrolysis process in the Ni cermet is only affected to a limited degree and independent on the sulphur concentration in the range from 6 - 14 ppm. The results show that the presence of sulphur reduces the steam reforming activity as expected from catalysis knowledge, and since the cell voltage degradation is only limited, and remain well below the thermo neutral voltage, this occurs without sacrificing too much of the electrochemical activity. In other words, up-grading of biogas using SOEC with Ni-ScYSZ electrode seems feasible.

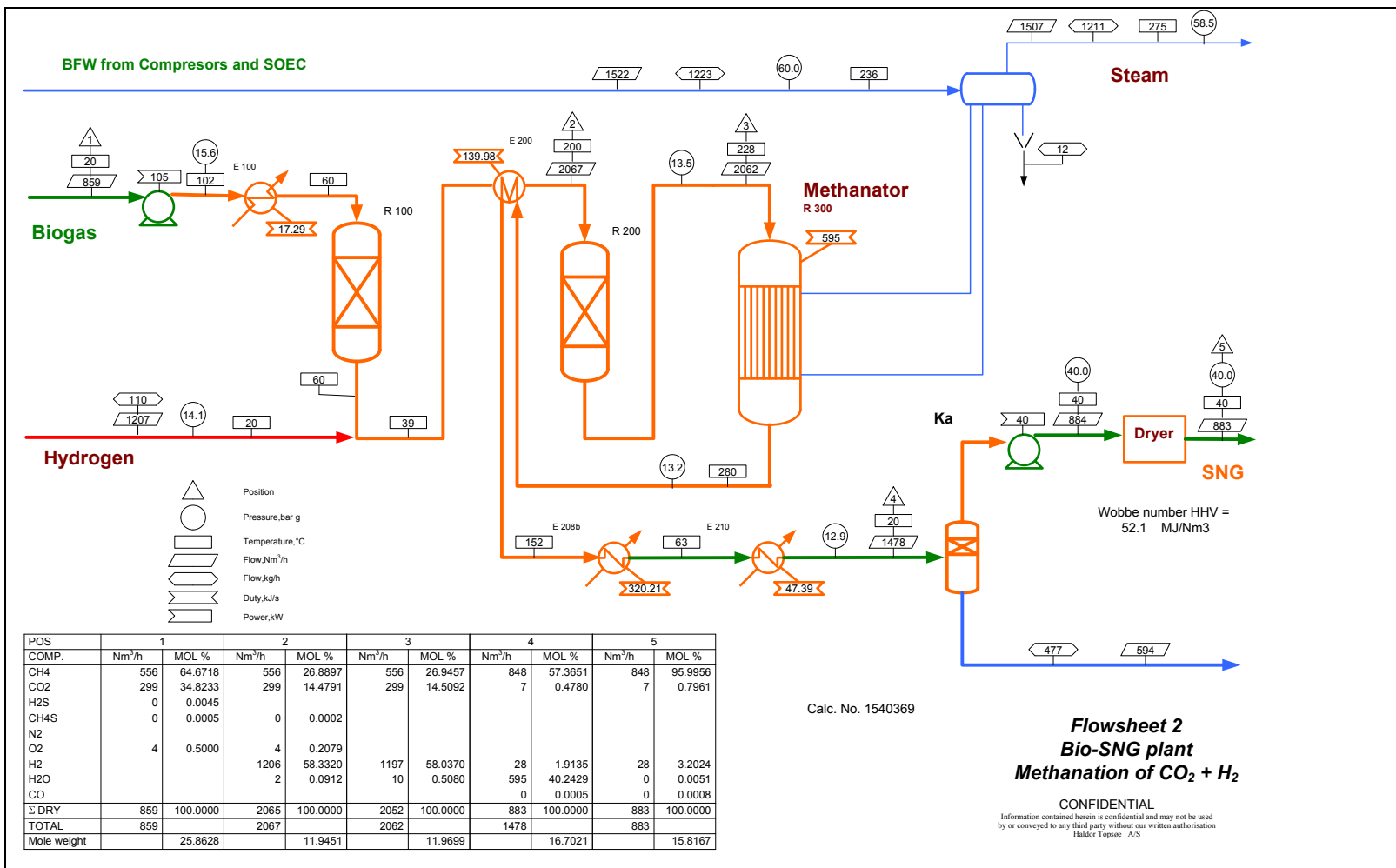
This finding makes it worthwhile to investigate the upgrading of biogas using SOEC much more in depth with the purpose of commercialising this technology. An increase of electrode performance and/or further development of new cells types such as the metal supported cell may help decreasing the costs. A proof of feasibility on stack level of say a few kW size will be important in order to reveal new possible challenges.

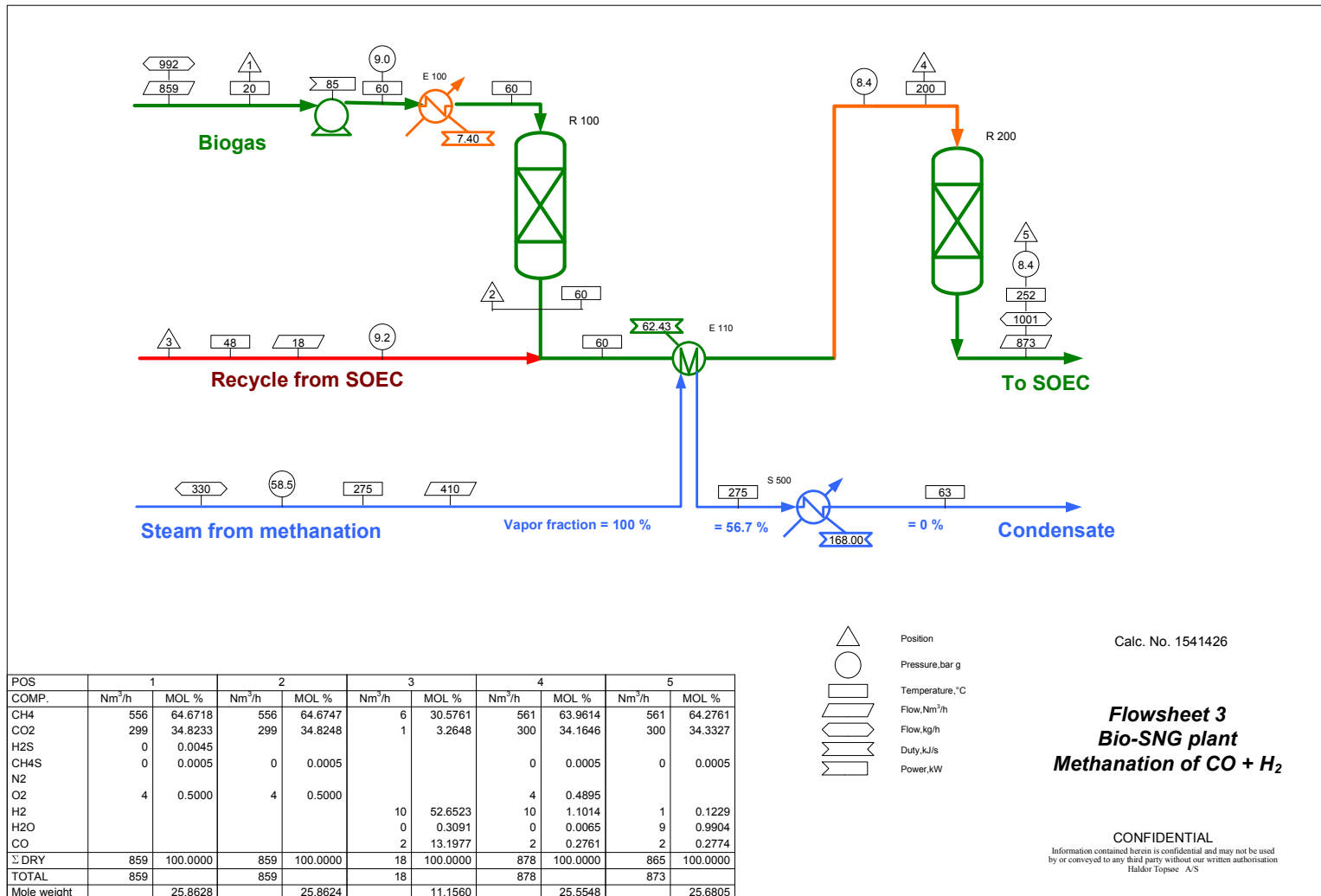
5 References

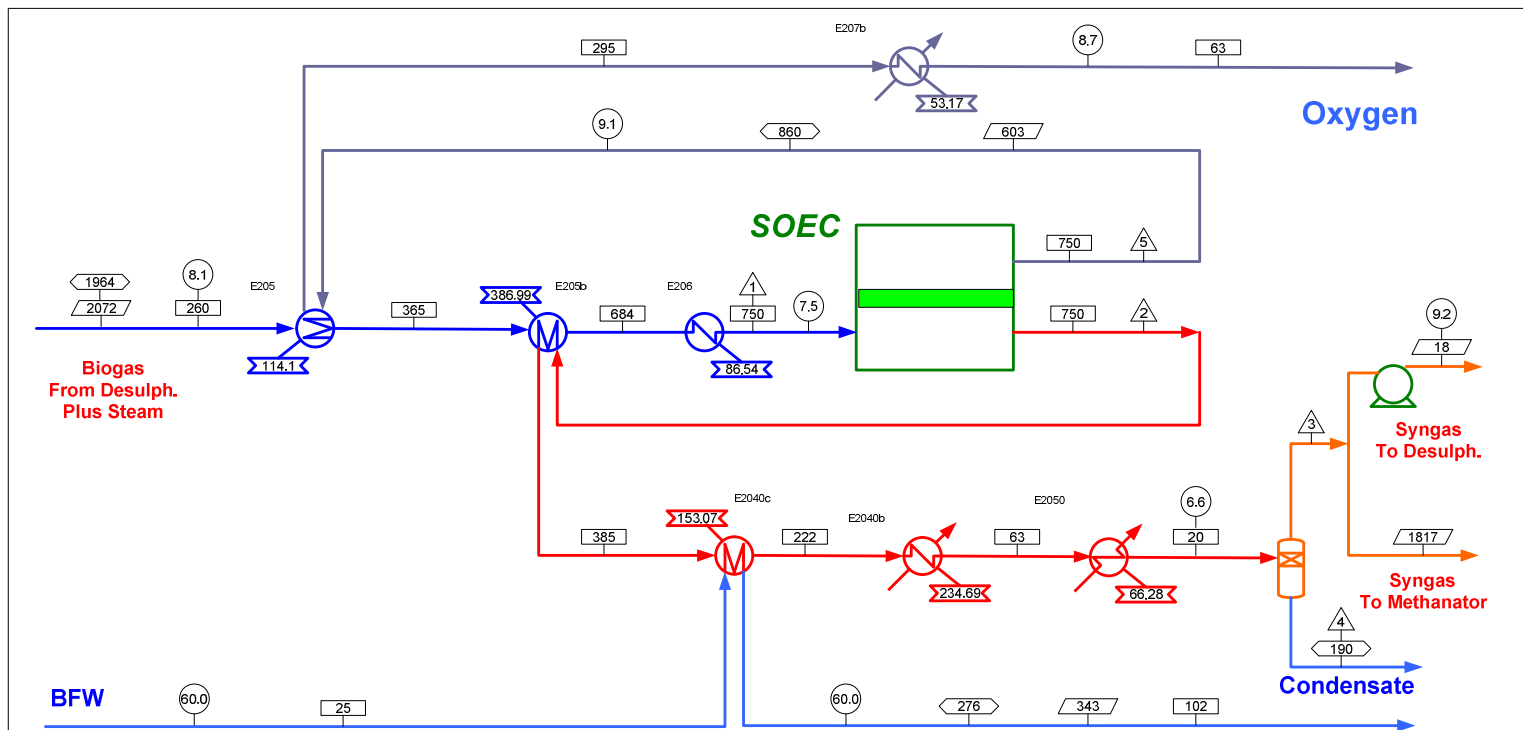
1. A. Hagen, J. F. B. Rasmussen, k. Thyden, *J. Power Sources*, **196**, 7271 (2011).
2. T. Klemensø, J. Nielsen, P. Blennow, A. H. Persson, T. Stegk, B. H. Christensen, S. Sonderby, *J. Power Sources*, **196**, 9459 (2011).
3. P. Blennow, J. Hjelm, T. Klemensø, S. Ramousse, A. Kromp, A. Leonide, A. Weber, *J. Power Sources*, **196**, 7117 (2011).
4. M. Mogensen, P. V. Hendriksen, in *High temperature solid oxide fuel cells - Fundamentals, design and applications*, p. 261 Elsevier, London (2003).
5. S. D. Ebbesen, C. Graves, M. Mogensen, *Int. J. Green Energy*, **6**, 646 (2009).
6. S. H. Jensen, A. Hauch, P. V. Hendriksen, M. Mogensen, N. Bonanos, T. Jacobsen, *J. Electrochem. Soc.*, **154**, B1325 (2007).

7. S. H. Jensen, *Solid Oxide Electrolyser Cell*, PhD Thesis, Risø National Laboratory, Roskilde, Denmark (2006).
8. S. D. Ebbesen , M. Mogensen, *J. Power Sources*, **193**, 349 (2009).
9. S. H. Jensen, A. Hauch, P. V. Hendriksen, M. Mogensen, *J. Electrochem. Soc.*, **156**, B757 (2009).
10. J. F. B. Rasmussen , A. Hagen, *J. Power Sources*, **191**, 534 (2009).
11. S. D. Ebbesen , M. Mogensen, *Electrochem. Solid-State Lett.*, **13**, B106 (2010).
12. S. D. Ebbesen, C. Graves, A. Hauch, S. H. Jensen, M. Mogensen, *J. Electrochem. Soc.*, **157**, B1419 (2010).
13. S. D. Ebbesen, J. Høgh, K. A. Agersted, J. U. Nielsen, M. Mogensen, *Int. J. Hydrogen Energy*, **36**, 7363 (2011).
















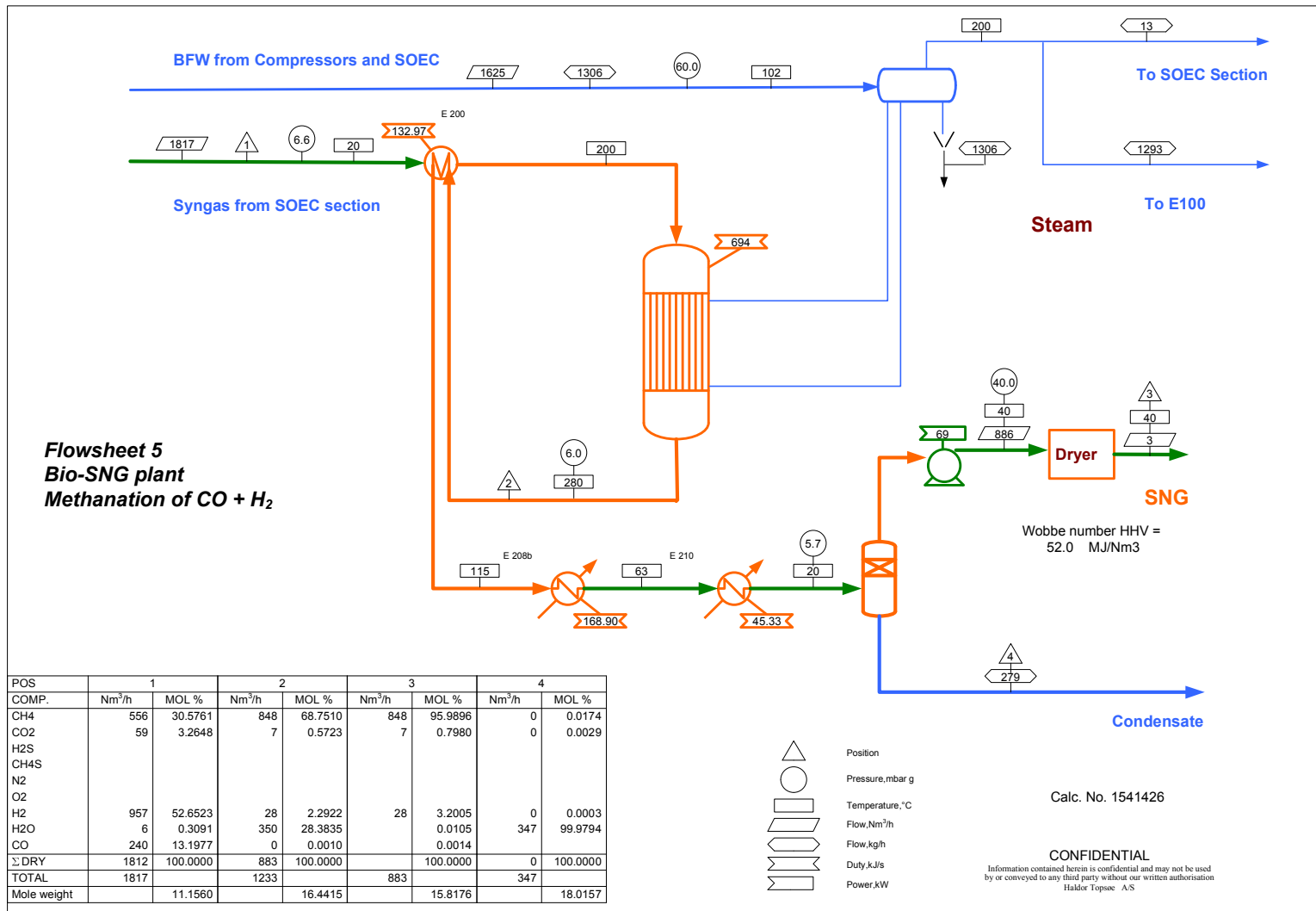
POS	1		2		3		4		5	
COMP.	Nm ³ /h	MOL %	Nm ³ /h	MOL %	Nm ³ /h	MOL %	Nm ³ /h	MOL %	Nm ³ /h	MOL %
CH4	561	27.0968	561	27.0968	561	30.5761	0	0.0063		
CO2	300	14.4736	60	2.8947	60	3.2648	0	0.0133		
H2S										
CH4S										
N2										
O2									603	100.0000
H2	1	0.0518	967	46.6604	967	52.6523	0	0.0060		
H2O	1207	58.2608	241	11.6522	6	0.3091	236	99.9725		
CO	2	0.1170	242	11.6959	242	13.1977	0	0.0018		
Σ DRY	865	100.0000	1830	100.0000	1830	100.0000	0	100.0000	603	100.0000
TOTAL	2072		2072		1836		236		603	
Mole weight		21.2467		11.9370		11.1560		18.0179		31.9988

-  Position
-  Pressure, bar g
-  Temperature, °C
-  Flow, Nm³/h
-  Flow, kg/h
-  Duty, kJ/s
-  Power, kW

Calc. No. 1541426

Flowsheet 4
Bio-SNG plant
Methanation of CO + H₂

CONFIDENTIAL
 Information contained herein is confidential and may not be used
 by or conveyed to any third party without our written authorization
 Haldor Topsoe A/S



Electrical Biogas Upgrade

Business & Development Plan



Table of contents

1	INTRODUCTION	3
2	THE ECONOMY	4
3	THE MARKET	5
4	THE OPPORTUNITIES FOR HALDOR TOPSØE A/S	6
5	SWOT	6
5.1	STRENGTHS.....	6
5.2	WEAKNESSES.....	7
5.3	OPPORTUNITIES.....	7
5.4	RISK.....	7
6	CONCLUSION AND PROPOSED ACTIONS	7
7	REFERENCES	8

1 Introduction

Biogas is a source of renewable energy with a wide range of advantages for the general society:

- Biogas can replace fossil fuels such as natural gas
- Biogas is produced largely from waste (e.g. manure and municipal waste) and hence reduces environmental issues elsewhere in the society
- Biogas reduces green house gas emissions not only by replacing fossil fuels but also by reducing potential green house gas emissions (e.g. methane) from the untreated feedstock.
- The residue from biogas can be used as fertiliser and helps preserve critical nutrients such as phosphor
- Biogas production creates jobs in rural areas

For these reasons substantial biogas subsidies were granted in the recent Danish “energi forlig” and similar subsidies are provided in a number of other countries such as for example Germany and Sweden

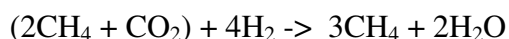
On this background, a large growth in the market for biomass production equipment is expected in the future. The present Danish production is expected to grow from presently 4 PJ to approximately 20 PJ around 2020 to achieve the political goal of using 50% of the manure for biogas production. Large growth rates are also expected world wide. In [1] it is estimated, that in an optimistic scenario, the 2 Mia € biogas plant installations of today may grow to 25 mia € in 2020.

In Denmark, biogas have been used mainly for local heat and power production (CHP), however with a substantially increased production, biogas supply will exceed CHP demand in many areas. In these cases it is relevant to upgrade the biogas, i.e. to remove the 30-40% of CO₂ in the biogas in order to obtain a methane rich (>96%) gas which complies with existing gas requirements and can be distributed via the natural gas grid.

.Presently only one biogas upgrade facility exists in Denmark but many more are expected in the future. In countries like Sweden, where biogas is used mainly for transportation and where the gas infrastructure is limited, biogas upgrade is widely used today.

Upgrade of biogas is however not free. Studies of biogas upgrade facilities in Sweden and Germany have concluded that the typical cost of biogas upgrade is in the order of 1 kr/Nm³ methane produced [2]. Rather than spending this relatively large amount of money on just removing the CO₂ from the raw biogas, several projects¹ have investigated the possibility of alternatively using the CO₂ for adding value to the biogas.

The idea here is to add hydrogen from electrolyzers to the biogas and use chemical synthesis to produce additional methane from CO₂ and hydrogen, i.e.



¹ In Denmark two feasibility studies have been carried out, “Methansamfundet” and “Bio-SOEC”. In Germany three facilities are planned, 1) SolarFuel is expected to start at 250 kW pilotfacility in 2012. 2) AUDI vil start to establish a 6,3 MW demofacility in 2013 and has planned for 1500 cars to run on ‘green gas’ from this facility. Erdgas Schwaben is considering a 1 MW demo facility

The advantages of this upgrade technique are that:

- Additional amounts of gas is produced which reduces either the need for import of energy or the use of limited biomass resources for energy production.
- Surplus electrical power is converted to gas, which can be stored in large quantities in the existing gas infrastructure (“Power to Gas”).

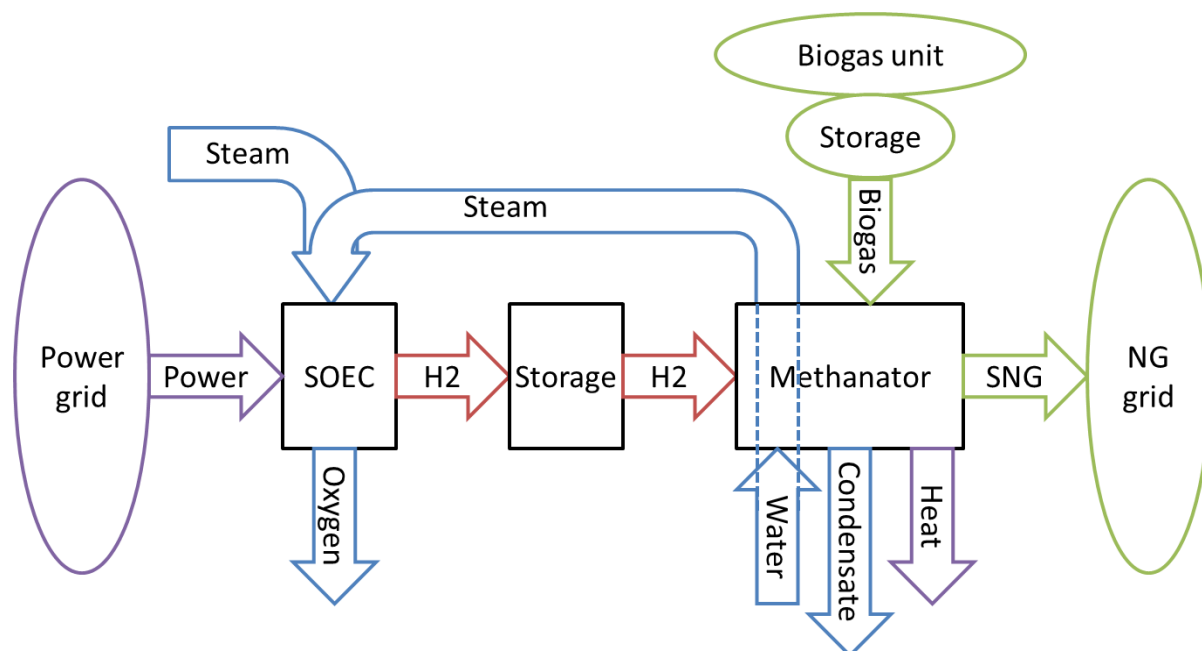


Figure 1. Possible process flow of electrical biogas upgrade[3]

In the general discussion on future energy system, “Power to Gas” is considered an attractive and perhaps even the only feasible approach to storing large amounts of intermittent electricity. This is particularly relevant in countries like Denmark and Germany where large quantities of intermittent renewable power has to be integrated into the power systems, in a not too distant future. The electrical biogas upgrade approach is a simple and relatively strait forward way of starting to introduce the power to gas technologies.

2 The Economy

Several Danish economical studies of the ‘electrical biogas upgrade’ scheme have been published as part of the two projects. “Methan Samundet” and “Bio-SOEC”. Here it is found that under the right circumstances the electrical biogas upgrade scheme can compete with the traditional CO₂-wash upgrade techniques.

For example in [4] it found that the cost of the methane produced from biogas CO₂ and electrolysed H₂ is 5 kr/Nm³, when SOEC is used as electrolysing technique. For comparison the cost of methane produced by traditional CO₂ wash of biogas is in the same study found to be 6.31 kr.

Assuming that the electrically upgraded biogas will be supported with the same subsidies as traditionally upgraded biogas, the business case for electrically upgraded biogas therefore looks quite favourable.

However, the electrical biogas upgrade business case is very uncertain:

- Electrical biogas upgrade is only viable if electricity can be obtained at spotprices w/o tax and transportation fees. This is not the case at the moment.
- Very efficient and cost-effective electrolysis techniques are required (e.g. SOEC) for the business to make sense. These electrolysis techniques have not yet been demonstrated outside the laboratory.
- Methanation is a well-known technique but the cost structure is only known for facilities which are orders of magnitudes larger than those needed for biogas upgrade.
- Using other economical assumptions the study in [3] finds that electrical biogas upgrade is not attractive in 2020 but will be in 2035
- In the two Danish economic studies [3] and [4], it is found necessary to operate the electrolyzers more than 8000 hours per year to obtain an attractive business case. Will it make sense for the society to support this technique if it can not help in load balancing the power grid? Alternatively, might it then be possible to reduce the cost of the electrolyzers to better support dynamic (<<8000 hours/year) operation?

3 The Market

According to [4] it is expected that approximately 50 new large biogas facilities with a total capacity of 500 mio Nm³ biogas per year will be established before 2020 in Denmark. Several of these will be placed in areas where local heat and power facilities are not expected to be able to use all the produced biogas (e.g. Lemvig, Skive and Samsø), in which cases biogas upgrade will therefore be needed. The numbers are uncertain but it is expected that from 10 to 50% of the new Danish biogas facilities will eventually include upgrading facilities. This gives a total Danish market for biogas upgrade of between 6000 and 30.000 Nm³/h corresponding to 150 to 750 mio kr for traditional upgrade and at least the double for electrical biogas upgrade.

Denmark has a substantial combined heat and power capacity and the need for biogas upgrade is therefore less in Denmark than in many other countries. In countries like Sweden and Germany, the market for biogas upgrade is already well established and growing rapidly. In [5] it is estimated that a German biogas upgrade capacity of 64.000 Nm³/h (methane) is installed today and that this will grow to 700.000 Nm³/h (methane) by 2020. This corresponds to a German biogas upgrade market of 2.5 mia kr from now to 2020 for traditional upgrade and potentially around 5 mia for electrical upgrade techniques.

Germany is presently the global market leader in biogas installations and the German market is estimated [1] to account for roughly 1/6 of the global market. By simple extrapolation it can therefore be estimated that the global market potential for the electrical biogas upgrade is of the order of 30 mia kr towards 2020 or of the order of several mia kr/year around 2020 when the technology is expected to be ready for large scale commercial launch. This is probably a very optimistic estimate as the upgrade market is expected to take up first in Europe and in particular in Germany[1]. The main point here is therefore not the number in it self but the fact that there is a large potential market, that the market eventually will be global and that the market will start in Europe most noticeably in Germany

So the potential market is definitely there, however these market estimates are extremely uncertain:

- Biomass is not expected to be able to compete price wise with natural gas. Government incentives are therefore required for large scale biogas facilities. These incentives have to be justified in local environmental concerns, CO₂ emission considerations or energy independence policies and may change with changes in the political agenda.
- The electrical biogas upgrade scheme is most relevant in regions with an ambitious policy for the integration of large scale solar- and wind-power. The situation here in year 2020 in different regions is very hard to predict.
- If the biomass can be used locally (for CHP), upgrading is probably not attractive unless there is a political incentive to use biogas for transportation. This will depend entirely on local conditions

4 The opportunities for Haldor Topsøe A/S

With a unique combination of technologies and market presence, the Haldor Topsoe group is well positioned to address the opportunities of the potential market for electrical biogas upgrade:

- Efficient electrolysis is the key to competitive electrical biogas upgrade solutions. Here SOEC is without competition the most efficient technology in particular in cases where waste steam is available as it will be in relation to methanisation. TOFC is one of the worlds leading companies in development and marketing of Solid Oxide cells, stacks and sub-systems.
- HTAS is one of the worlds leading companies in development and deployment of methanisation catalyst and systems.
- Our neighbouring countries Sweden and Germany are probably the two countries in the world where biogas upgrade facilities are most widely installed. Denmark has now some very attractive subsidies on biogas and a many new biogas facilities are expected in the coming years. Denmark and Germany are probably the two countries in the world with the largest need to test and willingness to invest in power to gas facilities.

So all in all this gives Haldor Topsøe A/S some unique opportunities in this potentially huge but also very uncertain market.

One particular issue for HTAS is that the present business model is build around supply of catalyst and engineering services. The catalysis needed for the biogas methanation will be very limited and the biogas upgrade business case does not allow dedicated engineering services at the individual sites. Consequently, it is not evident how HTAS is going to make money on this market. Supplies of SOEC systems are definitely an opportunity, but the company will also have to investigate business models for the entire upgrade facility.

5 SWOT

5.1 Strengths

- HTAS has a strong technical position with respect to SOEC and methanation technology
- Electrical biogas upgrade reduces CO₂ emission and dependence on foreign supply of fossil fuels
- Integration of SOEC and steam generation from the methanation provides unique efficiency

- HTAS is geographically close to the first market opportunities (Denmark, Germany and perhaps Sweden)
- There are good opportunities for testing and developing the technology with Danish partners and with Danish public support

5.2 Weaknesses

- SOEC has not yet been demonstrated outside the lab
- The necessary taxation scheme is not in place (no tax on electricity used for biomass upgrade)
- The upgrade business does not fit into HTAS existing business model
- The biogas market relies heavily on government subsidies
- Electrical biogas upgrade might only be profitable with >8000 hours of electrolysis per year implying very little potential for load balancing

5.3 Opportunities

- Electrical biogas upgrade might be the most cost-effective upgrade technology under the right circumstances.
- SOEC stacks have the possibility also to operate as fuel cell stacks in SOFC mode. This could provide an opportunity for more dynamic operation of the system and hence to give the desired load balancing capability of the electrical upgrade systems
- High pressure steam from the methanation might be used to pre-treat the biomass feedstock for higher biogas output
- Large scale biogas upgrade facilities will be established in the future, e.g. via Ringkøbing modellen. This will reduce the cost of electrical biogas upgrade

5.4 Risk

- The upgrade market might take off before the technology (mainly SOEC) is ready
- Alternative techniques for 'Power-to-gas' may be more attractive. This could for example be to insert 5-10% H₂ directly into the gas grid
- German competitors might be first to the market
- The biogas market may not take off as expected
- The proposed technologies are too expensive (SOEC and small scale methanation)

6 Conclusion and proposed actions

Electrical biogas upgrade is well aligned with important megatrends in the society and HTAS has some unique technical opportunities to address this market and HTAS is geographically close to the markets expected to emerge first. However, the business case of the electrical biogas upgrade is very uncertain both in terms of technology, legislation and market.

It is therefore proposed that HTAS starts small scale testing of the technology but does not start a full scale commercial development before some of the uncertainties have been addressed via field tests and commercial feasibility studies

7 References

[1]	GIA Industries White Paper, "How to Profit from Biogas Market Developments", June 2010, http://www.xebecinc.com/pdf/GIA_Industries_White_Paper_How_to_Profit_from_Biogas_Market_Developments.pdf
[2]	Energistyrelsen, "Opgradering og levering af biogas til naturgasnettet", Notat 2008
[3]	EA Energi Analyse A/S, "Biogas-SOEC, Energy system integration and economy" August 2012
[4]	Plan Energi,
[5]	Biogaspartner. Biogaseinspeisung in Deutschland und Europa. Markt, Technik und Akteure. 2011