Final report

1.1 Project details

Project title	Green Natural Gas		
Project identification (pro- gram abbrev. and file)	Journal No: 64011-0036		
Name of the programme which has funded the project	EUDP		
Project managing compa- ny/institution (name and ad- dress)	Haldor Topsøe A/S Haldor Topsøe Alle 1		
Project partners	 2800 Kgs Lyngby Haldor Topsøe A/S (project leader) DONG Energy A/S Dansk Gasteknisk Center A/S Technical University of Denmark, DTU Electro Technical University of Denmark, RISØ, Fuel cells and Solid State Chemistry department Ea energy Analyses 		
CVR (central business register)	Haldor Topsøe A/S: 41853816		
Date for submission	3/7-2015		

1.2 Short description of project objective and results

The objective of the Green Natural Gas project has been to demonstrate the feasibility of integrating the gas and electricity systems by producing green gas from CO_2 and renewable electricity.

The main results of the project are:

- Cost modelling based on parameters found in the project indicates that in the future it will be more economical to produce green gas from a combination of biomass and electrolysis rather than just from biomass
- A demonstration unit of a new and efficient electrolysis technology (SOEC) has obtained more than 890% power to gas energy efficiency

2.

Formålet med Green Natural Gas projektet har været at demonstrere muligheden for at integrere energisystemets gas- og elnet ved at producer grøn gas (metan) ud_fra CO2 og grøn eleektricitet

Projektets vigtigste resultater er at:-

- Kost modellering indikerer at <u>fra og med en gang mellem år 2020 og 2035 vilwill</u> det økonomisk være billigere at producer grøn gas_ud fra en ekombination af biomasse og eleektrolyseis end at gøre det alene ud fra elektrolyse
- Med en demonstrationsenhed af en ny elektrolyse teknologi (SOEC) er der opnået en energig effektivitet il omdannelse fra strøm til gas på over 90%

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1.3 Executive summary

In the Green Natural Gas project methane (Synthetic Natural Gas) will be produced from hydrogen and CO2, i.e.

CO₂ + 4H₂ -> CH₄ + 2 H₂O

This synthetic natural gas is referred to as 'Green Natural Gas' when the CO_2 used would otherwise be released into the atmosphere and when the power used for electrolysis is generated by renewable sources, i.e.

 $2H_2O$ (+green electricity) -> $2H_2 + O_2$

This project combines technology development with feasibility studies and cost modelling.

The main results of the project are:

- Cost modelling based on parameters found in the project indicates that in the future it will be more economical to produce green gas from a combination of biomass and electrolysis rather than just from biomass
- 2. A new an efficient electrolysis technology (SOEC) has been demonstrated. This has been done both in the form of a full system demonstration unit and by pressurised operation up to 25 Bar

In terms of commercial utilisation of the project results, Haldor Topsøe A/S has already moved quite far into the commercialisation of the SOEC technology. Based on the promising results obtained for the SOECCore in this project Haldor Topsøe decided to launch a commercial product for on-site (H2 and CO) production.

Such units would be placed in small containers and be sold to customers in the specialty gas business. The first of these containers (eCOs Plants) were <u>buildbuilt</u> in 2014 and the first unit has sold for delivery to a customer in 2015.

1.4 Project objectives

The objective of this project has been to provide systems analysis and key component development for 'Green Natural Gas' (Green-NG) production based on Solid Oxide electrolysis and renewable of CO₂ and electricity. By Green-NG is understood a 'green' methane (CH₄) based gas which is compatible with the existing natural gas grid. Solid Oxide Electrolysis (SOEC) is chosen as electrolysis technology because it is the potentially most energy- and cost-efficient electrolysis technology. The project has included a cost-analysis for Green-NG systems with the objective of pointing out the most promising paths forward for the technology.

The project activities have been directed towards feasibility studies, technology development, cost modelling and dissemination. The feasibility studies have included feedstock analysis, fuel synthesis concepts and gas grid integration. It is an objective to quantify the amount and cost of CO_2 potentially available for gas production. This analysis has been focussed on the DONGs biofuel facilities available at the start of the project: Inbicon, Renescience and biomass gasification.

The SOEC technology is a key element in this project and is expected to enable Green-NG systems which are about 33% more cost-effective than possible with other electrolysis technologies. In the project pressurised SOEC operation has been characterised as one important element in a cost-effective electrolyser design. Furthermore, an integrated SOEC submodule (SOECCore) is developed and tested as first critical step towards a future commercialisation of the SOEC systems technology.

The project is seen as an essential first step towards developing cost-effective Green-NG systems; however this project will only be one of several projects towards commercial Green-NG. In this project a 30 kW SOEC electrolyser was constructed and tested which provides important input to qualify the roadmap for the next development and demonstration phases.

1.5 Project results and dissemination of results

1.5.1 Solid Oxide Electrolysis

Solid oxide electrolysis (SOEC) has a range of unique characteristics which can make it a key element in a future CO_2 neutral energy system where fuels (e.g. green natural gas) are produced from renewable sources such as biomass and renewable electricity.

- Due to the high operating temperature (>700 °C), SOEC can have electrical to chemical energy conversion efficiencies close to 100%
- Being based on an oxygen membrane, SOEC can produce CO from CO_2 and H_2 from H_2O
- The same solid oxide stacks can be used in both fuel cell and electrolysis mode

1.5.1.1 Pressurised cells

One of the potential issues of SOEC is that due to the high operating temperature, it is more demanding to pressurize the cells and stacks than it is for traditional low temperature electrolysis technologies (PEM and Alcaline). This challenge arises from the lack of high-temperature gaskets which makes it necessary to build a pressure vessel around the stack rather than just pressurizing the stack.

The strategy for pressurized SOEC testing in this project has been to place the test setup into a pressure vessel that can accommodate a complete cell testing setup. By placing the whole test rig inside the autoclave, well-known and proven setups used for testing at atmospheric pressure can also be applied to high pressure testing.



Figure 5.1 Pressurised SOEC test rig

At DTU Energy two pressure test facilities have been constructed to test single solid oxide cells (SOC) and small 1 kW SOC stacks at elevated pressure. One of the test rigs is shown in Figure 5.1.

Figure 5.2a and b shows the effect of pressure on the cell performance in both SOFC and SOEC mode. The iV curves (a) and impedance spectra (b) were recorded at 1 and 3 bar respectively. A lower cell resistance at high pressure can be seen from both the polarisation characterisation as well as the impedance spectra.

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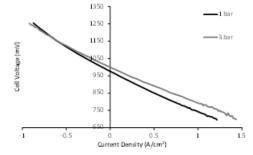


Figure 5.2.a: Polarisation characterisation in both SOFC and SOEC mode

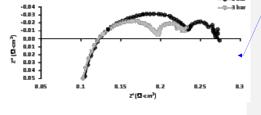


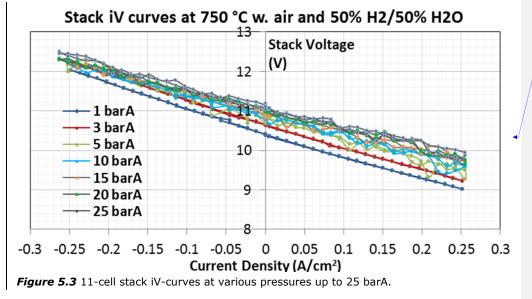
Figure 5.2 b: EIS at OCV at 800 °C with 50% H2 + 50% H2O supplied to the fuel electrode compartment and O2 supplied to the oxygen electrode

1.5.1.2 Pressurized stacks

iV-curves (Fig. 5.3) were recorded at various pressures on an 11-cell HTAS stack at 750 °C with 50% H_2 + 50% H_2O to the negative electrodes and air to the positive electrodes. The flow rates were 200 l/h H_2 + 200 l/h H_2O and 400 l/h air. The total electrode area in the stack was 965 cm², which gives an area specific flow rate of 0.41 l·h⁻¹·cm⁻². The H_2 and O_2 utilization at maximum current density in fuel cell direction was 46% and 54%, respectively. The H_2O utilization at maximum current density in electrolysis mode was also 46%.

The stack voltage was observed to fluctuate at increasing pressure. The evaporation temperature of H_2O increases with increasing temperature. This induced instabilities in the H_2O evaporation/condensation system which caused instabilities in the H_2O flow which again affected the stack voltage. During the autumn 2015 the H_2O evaporation/condensation system will be improved to remove the stack voltage fluctuations at elevated pressure.

The open circuit voltage (OCV) (i.e. the stack voltage at 0 A/cm²) is seen to increase with increasing pressure. Further, the slope of the iV curves is seen to decrease with increasing pressure, indicating that the internal area specific resistance (ASR) of the stack decreases with increasing pressure. Both the increase in OCV and the decrease in ASR is in agreement with the single-cell measurements presented in Fig. 5.2, theory and previously observed on single-cells at pressures ranging from 0.4 barA to 10 barA.



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1.5.1.3 Systems

Prior to this project, the different characteristics of SOEC had been demonstrated in R&D- and university-labs on a relatively small scale. An important ambition of this project was to perform a

large scale demonstration. Specifically, the goal was to demonstrate high efficiency on a 30 kW system.

Because of the high temperature operation of the SOEC stacks it was decided to divide the 30 kW SOEC system demonstration unit developed in this project into a 'hot part' and a 'cold part'.

The hot part of the system is called the SOEC Core and contains the stacks and the heat exchangers and electrical heaters used for the thermal management of the inlet and outlet gasses. The block diagram of the SOEC Core is shown in figure 5.4a and the mechanical layout is shown in figure 5.4b. Each core contain up to 4 stacks, where each stack has 75 cells.

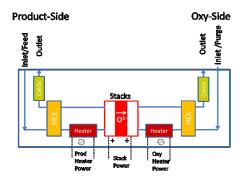


Figure 5.4a. The block diagram of the SOEC Core

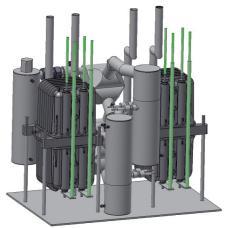


Figure 5.4b. The mechanical layout of the SOEC Core

The cold part of the system is called the eCOs rack and is (together with the SOEC-Core) shown in figure 5.5. The rack contains the instrumentation, the electronics and the safety system, e.g ventilation etc.

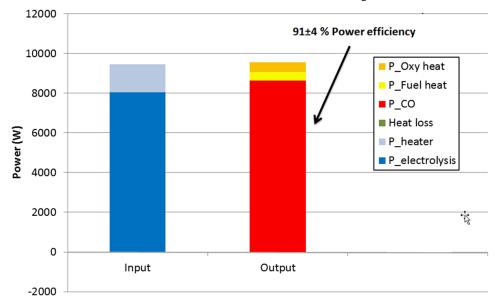
On figure 5.5 to the right the SOEC Core is the metal box in the center of the picture behind the center door.



Figure 5.5 The eCOs Rack and the SOEC Core

Figure 5.6 below indicates the power efficiency of the SOEC core when operated in CO_2 electrolysis mode. The stacks are operated electrically in series two and two and figure 5.6 summarized the energy balance for a series of two stacks when operated at 40A. Power is provided to the core by the DC-power supply (P_electrolysis = 8 kW) and the electrical

heaters (P_heater = 1.6 kW). This is shown in the blue column.



Power overview, Core3, 4 Nm³/h CO₂, 40 A

Figure 5.6 The energy balance for a series of two stacks in the SOEC Core when operated at 40A.

In figure 5.6 Energy is removed from the system by chemical energy (P_CO, 2.4 Nm3/h CO @ 3.5 kWh/Nm3 = 8,4 kW) and hot gasses at the oxy and fuel outlet sides. The thermal energy in the outlet gasses (P_Oxyheat and P_Fuelheat) reflects the energy loss of the system and are in this case 9% of the energy input, corresponding to a net efficiency of 91% for the core. For the entire system the efficiency is roughly 5% lower as there is some electrical loss related to the DC-power supplies and to the operation of instrumentation and ventilation.

1.5.2 Feasibility studies

1.5.2.1 CO2 Sources

 CO_2 is a critical feedstock for the production of SNG. CO_2 can be made available from a range of different sources and the capacity (Tons/Year) and cost of access/purification of the most important Danish sources have been mapped in this project.

- Several million tons of CO₂ per year could be made available by extracting it from the fluegas of combustion processes for power plants and cement production. For fluegas, the cost of CO₂ sequestration (removal of oxygen and nitrogen) is typically relatively expensive (100 €/ton).
- 10-100 thousands of tons of CO₂ can be made available from 'green sources' such as breweries, biogas and bioethanol production. For biogas sequestration cost are relatively high, where they are lower for breweries and bioethanol production. However, the amount of CO₂ available from the two latter sources is quite limited.

Major CO ₂ sources in	Tons CO ₂ /year	Price per tons of captured
Denmark		CO ₂ . (Sequestration cost)
Power production - CO ₂	17,5 million (from 9 major plants)	80 - 100 €/t to day (2012)
		30 - 40 €/t in 10 - 20
		years
Cement production	1,4 million (Ålborg Portland)	80 - 100 €/t
Breweries (Green)	6000 tons (De Forenede bryggerier,	25 - 30 €/t (not for sale)
	Fredericia)	
Biogas (Green)	3 - 6000 tons (per plant for app. 30	*) No CO ₂ production to
	plants) + 50.000 tons at Måbjerg	day.
	(2016)).	Possible capture cost -

		100 €/t	
Bioethanol (Green)	70.000 tons (10.000 in Kalundborg + 60.000 at Måbjerg (2016))	25 - 30 €/t (expected)	
Table 5.1 Summary of Danich CO, sources estimated in 2012			

Table 5.1. Summary of Danish CO₂ sources estimated in 2013

Sequestration cost of 100 \in /ton for CO₂ will add at least 1.5 kr/Nm³ to the cost of SNG synthesis (CO₂ + 4H₂ -> CH₄ + 2H₂O). Consequently, it is attractive to consider methanization of lower cost sources of CO₂. This could for example be non-purified sources of CO₂, where sequestration cost would be negligible. The two most attractive sources of non-purified CO₂ are:

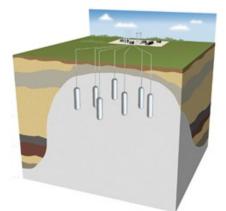
- Biogas, which typically contains 40-60% CO₂ mixed with CH₄
- Gasified biomass which typically contains 30% CO₂ mixed with H₂ and CO

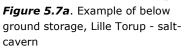
Alternatively, bioethanol could provide a relatively cost-effective source of pure CO_2 . Based on this feedstock analysis, the three cases considered for cost analysis in this project are electrolysis assisted Green Natural Gas based on CO_2 from 1) Biogas, 2) Gasified biomass, 3)Bioethanol

1.5.2.2 Gas storage

Gas storage can potentially be a critical technology to decouple the hydrogen production (electrolysis) from the methane production (gas synthesis). Ideally electrolysis should be used mainly when the electrical grid power supply exceeds power demand and electricity prices are low. This leads potentially to a very fluctuating electrolysis operation, where as it typically will be desirable to have a much more continuous operation of a gas synthesis plant.

In the project the availability and cost of a range of different above- and below-ground storage technologies have been investigated. Two of these are illustrated in figure 5.7 below.





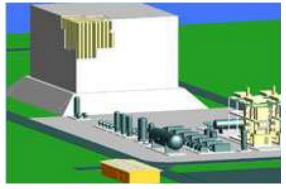


Figure 5.7b. Example of above ground storage: Vertical pipe storage solution

The gas storage cost estimates found in this project are summarised in table 3.2 for capacities considered relevant for the gasified biomass case. Here the a storage capacity correspondings to 48 hours of gas consumption.

	Cost per Nm ³ of gas storage €/Nm ³ gas		
Gas	Hydrogen	CO ₂	0 ₂
Capacity	10,5 MNm ³	0,5 MNm ³	1,5 MNm ³
Aquifair storage at the Havnsø	10,3	71,4	35,6
Existing Salt Caverns	2,9	31,4	16,9
New Salt caverns	3,9	51,4	23,5
(Vertical) pipe storage	20	61	38
Gas bottles	19	14	23

Table 5.2 Cost of gas storage for 48 hours of storage for the gasifed biomass case. The grey lines are for below ground storage and the blue lines for above ground storage

Based on these results it is concluded that:

- Large volumes of H₂ can be stored quite cost effectively in large caverns. This will of course limit the relevant locations available for the green natural gas plant
- In terms of storage cost per Nm³, CO₂ and O₂ storage is significantly more expensive per than storage of hydrogen. In particular the cost of O₂ storage is difficult to estimate as a number of additional cost may arise from measures needed to protect against oxygen fuelled fire.
- The required storage volume of hydrogen is much larger than for CO₂ and O₂. Consequently, the cost of hydrogen storage is the dominating storage cost (not including the possible cost of storing the produced CH₄)

1.5.2.3 Gas quality

In Denmark natural gas has to conform to the National Gas Regulations in order for the gas to be admitted to the natural gas system. In the Gas regulation it is specified within which range of the Wobbe-index and relative density the gas should conform. At present the range for the Wobbe-index is 50.8 to 55.8 MJ/Nm^3 (at 0 °C) and the range for the relative density is 0.555 to 0.700. The relation between the heating value, the relative density and the Wobbe index is calculated as:

Wobbe index = $\frac{Superior Heating Value}{\sqrt{Relative Density}}$

The limits for the present regulations are plotted in figure 5.8. The black dotted line show the limits for the Wobbe-index and the red and blue solid lines shows the limits for the relative density.

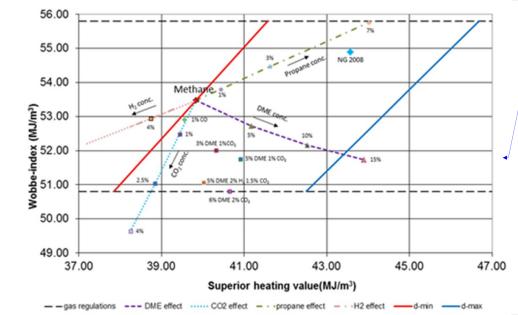
Pure methane only just complies with the regulation, it would therefore most likely be necessary to add a component which can move the SNG into legal area:

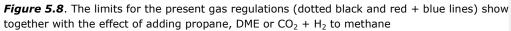
- 1. For upgraded biogas, propane is often used today. It is however desirable to avoid the use of fossil additives to a 'green gas'
- 2. In the project, it was considered to add DME and this is shown in figure 5.8 with the purple line. This will increases the heating value but reduce the Wobbe index.
- For methane synthesized from CO₂ and H₂, there will be some remaining feedgasses in the product. With these gasses remaining in product the gas 'quality' will move 'downwards' in the diagram as indicated with the blue dotted line. If for example 2.5% of CO₂ remains in the gas, up to 1.6% of H₂ can be tolerated.

It is estimated that sufficiently pure methane can be synthesized from CO_2 and H_2 to meet the requirements of bullet 3 and in the cost modelling of the different green natural gas scenarios it has therefore been assumed that no additional gasses would have to be added.

It would probably make future green natural gas system simpler if lower Wobbe indices could be tolerated. In the European "Gas quality Harmonisation Pilot Project" new cross-European gas regulations are presently been discussed. Here it has been proposed, to extend the limits for the Wobbe index to 48.5-56.9 MJ/Nm³ (at 0 °C). This would simplify the introduction of Green Natural Gas, however these discussion are still at an early phase.

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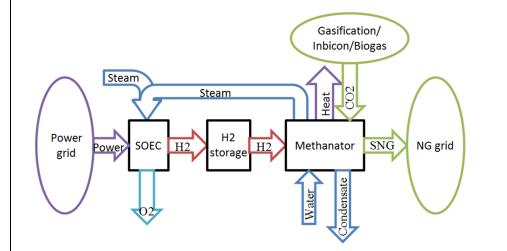
1.5.3 Economic Modelling

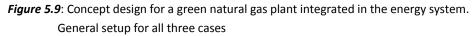
The purpose of the economic modelling of the three green natural gas scenarios have been to compare the cost of Methane gas produced from a combination of biomass and SOEC with the cost of Methane produced from just biomass.

An economic optimization model was constructed to simulate the Synthesized Natural Gas (SNG) prices and profitability of three different SNG production plants in the three years 2020, 2035 and 2050. The three different plants are listed in the table below and the general system outline is shown in figure

	Bio based CH ₄ (NM ³ /h)	SOEC based CH ₄ (Nm ³ /h)	External reference
Gasified biomass	20.000	12.500	Bio2G (E.ON Swe- den)
Biogas	575	300	
Bioethanol	0 (Pure CO2)	2020: 240 2035: 600 2050: 3600	Inbicon, Kalundborg Inbicon2, Kalundborg Mårbjerg

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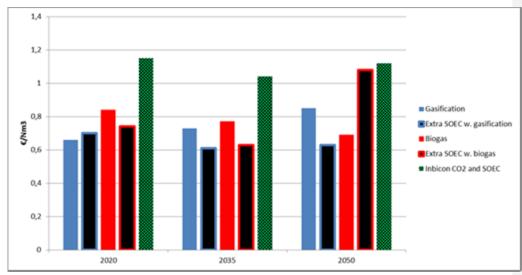


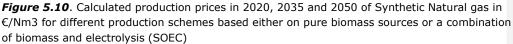


Calculating the absolute price ad profitability of the Synthetic Natural Gas produced (SNG) will of course depend on a wide range of parameters of which most become quite uncertain as we move beyond 2020. Some of this uncertainty can be reduced by comparing SNG produced from biomass with SNG produced from biomass and SNG. The assumption here is that it is politically decided that SNG (or other CO_2 neutral fuels) are needed and the question is then if it is a good idea to add electrolyzed hydrogen and methanisation to the process. In this case the main input parameters to be estimated are:

- Electricity prices: By using the Balmorel model, electricity prices series for the three years 2020, 2035 and 2050 was simulated. The Balmoral model utilizes a mix of 'green' and 'black' feedstocks for producing electricity. In 2035 77% of the power used is 'green' and this increases to 97% in 2050.
- 2. SOEC cost, lifetime and efficiency. Here an SOEC cost price of 1000 €/Nm/h were used, a lifetime of 5 years lifetime and an efficiency of 95%
- 3. Cost of hydrogen storage and CO₂ were as described in section 3

In this case, the economic modeling results shown in figure 5.10 indicates that from 2035 it is more cost-efficient to produce Green Natural Gas by a combination of SOEC (and green power) & gasification than by either of the two technologies on their own. Please be aware that the SOEC prices used in the simulations are not expected to be reached in 2020 and that the cost estimates for SOEC produced gas in 2020 probably are a bit too optimistic.





A critical parameter in these simulations is the fluctuations in the electricity prices as larger fluctuations will make the use of SOEC more profitable. Comparing with actual fluctuations in the power prices, it is well known that the Balmoral model have has a tendency to underestimate the power price fluctuations. For the Balmoral simulations used here the fluctuations (standard variation) in the power are predicted to be lower in 2020 and 2035 than they were in 2013. This not a like scenario as power price fluctuations typically increase as the ratio of renewable energy increases in the grid. To address this the following simulation steps were taken:

- Using historical (2013) electricity price fluctuation levels (standard variation) for the simulated years gives a slightly lower (2%) price of SOEC based gas in 2020 and 2035, while the price is 10% lower in 2050.
- Fluctuations are expected to increase in the future as the amount of renewable energy in the grid increases. Assuming a fluctuation level (standard variation) which is 50% higher than in 2013, the SOEC based gas prices are 7-15% lower in 2020 and 2035, while the extrapolations does not provide meaningful results in 2050

1.5.4 Dissimation of results

1.5.4.1 Peer-reviewed journal publications:

Sune D. Ebbesen, Xiufu Sun, and Mogens B. Mogensen. Understanding the processes governing performance and durability of solid oxide electrolysis cells. Faraday Discussion, 2015, DOI: 10.1039/c5fd00032g

Xiufu Sun, Alfredo D. Bonaccorso, Christopher R. Graves, Sune D. Ebbesen, Søren H. Jensen, Anke Hagen, Peter Holtsappels, Peter V. Hendriksen, and Mogens B. Mogensen. Performance characterization of solid oxide cells under high pressure. Fuel Cells, 2015, Accepted

Sune D. Ebbesen, Søren H. Jensen, Anne Hauch, and Mogens B. Mogensen. High Temperature Electrolysis in Alkaline Cells, Solid Proton Conducting Cells, and Solid Oxide Cells Chemical Reviews, 2014, 114, 10697 – 10734

1.5.4.2 Peer-reviewed proceedings publications:

Xiufu Sun, Alfredo D. Bonaccorso, Christopher Graves, Sune D. Ebbesen, Søren H. Jensen, Anke Hagen, Peter Holtappels, Peter V. Hendriksen, Mogens B. Mogensen. Performance characterization of solid oxide cells under high pressure. Proceedings of the 11th European SOFC & SOE Forum, 2014, B1301, 5 – 13.

1.5.4.3 Other publications

Niels Bjarne Rasmussen, "Nye gaskvaliteter på naturgasnettet", Gasteknik Nr5, 2014

Aksle Hauge Petersen, Koen G. Wiersma, "Renewables: Technological and economic aspects of power to gas and upgrading of biogas to natural gas quality", IGU World Gas Conference 2015, Paris

Koen G. Wiersma, Aksle Hauge Petersen, "Making the case for Power-to-Gas", IGU Magazine April, 2015.

1.5.4.4 Conference presentations

John Bøgild Hansen, 13 maj, 2012: Danske Gastekniske dage, Billund

John Bøgild Hansen, 27 April 2013, Brazilean CO2 conference, Rio de janeiro

John Bøgild Hansen, 10 marts 2014 Power to Gas Conference, Düsseldorf

John Bøgild Hansen, 27 august 2014: Nordisk Biogas konference, Reykjavik

1.6 Utilization of project results

In terms of commercial utilisation of the project results, Haldor Topsøe A/S has already moved quite far into the commercialisation of the SOEC technology. Based on the promising results obtained for the SOECCore in this project Haldor Topsøe decided to launch a commercial product for on-site (H_2 and CO) production.

Such units would be placed in small containers and be sold to customers in the specialty gas <u>market</u>business. The first of these containers (eCOs Plants) were build in 2014 and the first unit has sold for delivery to a customer in 2015.

It is expected that the eCOs products will help to establish a profitable SOEC stack and system business in the next couple of years. Furthermore, the commercial deliveries for the specialty gas market will play a very important role in driving SOEC cost and durability towards the targets needed to introduce SOEC into future energy system.

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Figure 6.1 The eCOs plant utilising SOEC technology to produce up to 15 Nm3/h of on-site gas for the specialty gas market.

1.7 Project conclusion and perspective

The economic modeling indicates that from 2035 it will be more cost-efficient to produce Green Natural Gas by a combination of SOEC (and green power) & gasification than by either of the two technologies on their own.

This require a significant development of the SOEC technology and the Green Natural Gas project has played a very important role here in supporting the development of the SOECCores, which has now enabled Haldor Topsøe to introduce SOEC systems commercially. The next critical steps to develop the SOEC technology and hence to enable Green Ntural Gas systems are believed to be:

- SOEC stacks have presently been tested for up to one year, while 5 years are needed to match the economic modelling. Increasing lifetime require further testing combined with improvements on cell and interconnect lifetime.
- The stack cost used in the model is roughly 1 k€/Nm3/h which is about one order of magnitude lower than present commercial SOEC stack costs. Technologically, such a price reduction is possible, but it requires much larger volumes produced than today. It is therefore essential to develop commercial SOEC applications which can drive volumes up and prices down toward the level needed for the introduction of SOEC into green natural gas systems. Such application can be for the specialty gas market (onsite CO and H₂) and possible also for off-grid energy storage (reversible SOEC/SOFC operation)
- This project has demonstrated the efficiency of SOEC at the system level and pressurisationpressurization at the stack level. It is also important to demonstrate the other key aspects. In particular
 - The use of Reversible (SOEC/SOFC) systems is an important feature which potentially could reduce the cost of introducing SOEC in Green Natural Gas systems
 - o <u>Pressurisation</u>Pressurization at the system (SOECCore) level

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