Final report

1.1 Project details

Project title	Robust HT-MEAs for Dynamic Operation under Smart Grid Conditions (SmartMEA)
Project identification (program abbrev. and file)	Project number: 2014-1-12218
Name of the programme which has funded the project	ForskEL
Project managing company/institution (name and address)	Technical University of Denmark Department of Energy Conversion and Storage (DTU Energy) Kemitorvet 207, 2800 Lyngby
Project partners	DTU Department of Electro Technology (DTU Elektro) Danish Power Systems (DPS)
CVR (central business register)	30 06 09 46
Date for submission	17-12-2017

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1.2 Short description of project objective and results

1.2.1 Technical results

The main outcome of the project are directly or indirectly within cost reduction and cell durability. These are the most important areas for development of the rather mature High-temperature PEM fuel cell (HT-PEMFC) technology today.

On cell level the electrode structures have been studied and the understanding of the acid loss mechanism is expanded. I was shown that the choice of electrode support (the gas diffusion layer, GDL) strongly influences the rate of phosphoric acid loss and that the recommended material appeared to be the one of the four material studied that had the poorest acid retention capability. Elemental mapping gave new insight into the behaviour of the acid in the cell.

Electrospinning of electrode backing structures, i.e. for GDLs was initiated. Fibrous structures much finer than in the usual commercial GDLs were manufactures. However these materials still remain to be tested in fuel cells, and this will happen in the near future.

A new membrane technology was developed and pronounced reduction of the voltage decay rate was demonstrated over 10,000 h at 180 °C. the higher temperature was chosen to accelerated testing. Yet degradation rates as low as 2.7 and 4.5 µV h^{-1} were recorded over most of the test period. This is very low at 180 °C. The new technology is in the process of being patented.

The platinum loading was reduced by one third by the application of an alloy catalyst.

Dynamic testing of the single cells including load change and start-stop was initiated.

DTU Elektro carries out an extended study of the use of HT-PEMFC in the energy system.

1.2.2 Utilization

The findings of the project will be utilized for both the manufacturing of cells and cell components at DPS and for further scientific research at DTU Energy.

The technical development and knowledge gained will be of great importance to the company. The nature of the accomplishments of the project is technical and thus highly relevant to the general gearing up of DPS. The findings will certainly be utilized in the commercialization of HT-PEMFC for which DPS is a leading player.

At DTU Energy the results and new ideas of the project has triggered new pathways for research and development that will be pursued. The work on phosphoric acid movement and retention resulted in quite unexpected findings and a new Master project will be initiated together with DPS on the understanding of the activation process in the cells. Electrospinning is a hot topic internationally and in the project we have started several activities without being able to take it all the way to cell tests yet. This will a major activity at DTU Energy in the near future. Two new PhD projects and a postdoc project on electrospinning will start early 2018 at DTU Energy.

The project has helped CEE/DTU Elektro into a closer and better collaboration with DTU Energy, e.g. leading to a successful proposal in the H2020 and EUDP programs. Results from the project has led to increased CEE/DTU Elektro activity in the field of integrated energy systems. As consequence CEE has successfully engaged in new activities in the field between electric power, heat and gas. CEE has currently several PhD students working with integrated energy infrastructure.

1.3 Project objectives

1.3.1 Purpose of the project

The project core is research within and development of proton exchange membrane fuel cells (PEMFC) operating at elevated temperature - as compared to that of the most common type. The project title “SmartMEA” refers to the term MEA which is an abbreviation for “membrane electrode assembly” which is the core of each fuel cell in any PEMFC single cell or cell stack. “Smart” naturally refers to smart grid operation.

1.3.2 Expected findings

• Better understanding of the specific role HT-PEMFC can play in a future Smart Grid. Advantages/disadvantages. Benefits from the high temperature heat in a district heating system etc.

• More robust cells capable of dynamic operation in a Smart Grid

• Improved acid retention in the fuel cell

a) By modification of the electrodes

b) By improvements of the assembly method used

• Significant lowering of the MEA resistance coming from the electrode – membrane inter-face

• Improved sealing materials to make cells compatible with modern stacking techniques.

• DTU ELECTRO will obtain expertise in the HT-PEMFC technology, which is expected to be important for future activities of implementing the fuel cells

1.3.3 Objectives

Simulation and modelling

• Definition of dynamic operation modes of MEAs

• Simulation of fuel cell based electric power system and its fitting into smart grid

• Investigation of interplays of fuel cells and smart grids

• The specifications of MEA operational parameters and performance targets

• Input to the definition of MEA test protocols

Electrode development

• Development of more robust electrodes

• Immobilization of (solid/inorganic) proton conductors

• Development of heterogeneous structures of multilayers or/and electrospun materials

• Reduction of the MEA resistance in the electrode membrane interface.

• Improvement of phosphoric acid retention in the MEA

MEA Enforcement

• Studies and optimization of MEA compression

• Optimization of membrane edge reinforcement

MEA manufacturing

• Optimized MEA assembly to reduce acid redistribution

• MEA stacking using EWII developed stack for testing.

Dynamic tests

• To perform fuel cell evaluation of materials and MEA techniques under dynamic operation with objectives of

• Establishment of dynamic fuel cell testing protocols

• Further improvement of overall MEA performance by 50 mV at current density of 200 mA/cm^2

1.4 Project results and dissemination of results

The description of activities and technical results is done below work package by work package followed by reporting of dissemination.

WP 1. Smart Grid specifications

WP 2. Next generation electrodes

WP 3. Enforcement technologies

WP 4. MEA manufacturing

WP 5. Test protocol

Dissemination

1.4.1 WP 1. Smart Grid Simulation and MEA Specifications

(Lead: DTU Electro)

The following are an extract of the report “Robust HT-MEAs for dynamic operation under smart grid conditions”, 2017, hereafter referred to as *the report* . In particular chapter 3 (simulations) and 4 (parameters for fuel cells) applies with regards to simulation and fuel cell requirements/specifications for grid operation.

1.4.1.1 Simulation of cogeneration in Danish households

Fuel cell for cogeneration in households is simulated with respect to the operating energy costs of a typical Danish household during one year.

1.4.1.2 Methodology

In this study, the heat and power supply to a typical Danish household were modelled by means of traditional technologies and cogenerative fuel cells running on reformate hydrogen. The objective was to determine potential savings from the latter in the energy cost.

The most commonly deployed technologies are compared to fuel cells under different operating strategies. The following technologies were considered for which technical parameters are given in the report: district heating, gas boilers, heat pumps and HT-PEM fuel cells.

The total energy cost is calculated as the summation of the costs for utilities (grid power, fuel, and district heat) to the net of electricity sellback revenues when cogeneration is involved.

The dispatch strategy for the cogeneration case prioritizes auto production consumption over utilities purchase and varies as follows:

A. Heat-led operation

The fuel cell follows the heat demand at any given time (no thermal storage needed)

B. Power-led operation

The fuel cell follows the power demand at any given time and heat is stored if not immediately consumed. When needed, a backup boiler is used to meet the heat demand.

C. Partial power-led operation

The fuel cell partially covers the power demand at any given time and heat is stored if not immediately consumed. When needed, a backup boiler is used to meet the heat demand.

When relevant, two cases were assessed for gaseous fuel consumption, namely natural gas and bio-methane. This allows to consider the difference in the utility cost and to include the generous incentives for sellback of biogas based electricity currently sold at 145 €/MWh. Power sellback from natural gas fuel cells was settled at 30 €/MWh_{el} which is approximately the average spot price .

Natural gas is purchased at the average retail price for households in Denmark of 76 €/MWh. Grid electricity, bio-methane, and district heat prices were set at 300 €/MWh_{el}, 69 €/MWh_{th}, and 89 €/MWh_{th} respectively. Because the power demand profile was calculated for year 2016, the spot price time series were used for the same period.

Demand data available in literature (table 7 & 8 in the report) was used and typical hourly heat demand profiles derived from measurements taken on Bornholm Island. The electricity demand profile was that of West Denmark in 2016. The actual consumption per household was then calculated by fitting the profiles to the total consumption.

The energy demand includes electricity and heat (for both domestic hot water and space heating). The total heat demand was calculated by assuming that the average floor area is 147m^2 (ref 62 in the report) and the specific heat demand is 110 kWh/m^2 (ref 63 in the report) and was assumed to be 4 MWh$_{el}$, as in (ref 12 in the report).

1.4.1.3 Results

The total energy costs for each case are given in Figure 1. Traditional systems based on DH, boilers and HPs give total costs ranging approximately between 2600 €/year and 2800 €/year averaging about 2700 €/year. For cogenerative fuel cell systems the range is significantly wider, starting from 1900 €/year and up to 3000 €/year. When the fuel cell is used to meet the power demand (totally or partially, as in case B and case C respectively), the average energy cost is lower (2200 €/year on average) to that of traditional heating systems.

Opting for bio-methane over natural gas always returns lower total costs mainly due to their price difference (69 €/MWh$_{th}$ vs. 76 €/MWh$_{th}$) and to dedicated subsidies for biogas based power (145 €/MWh). However, the choice of bio-methane may be limited by availability and for most end users natural gas is likely to be the only possible option.

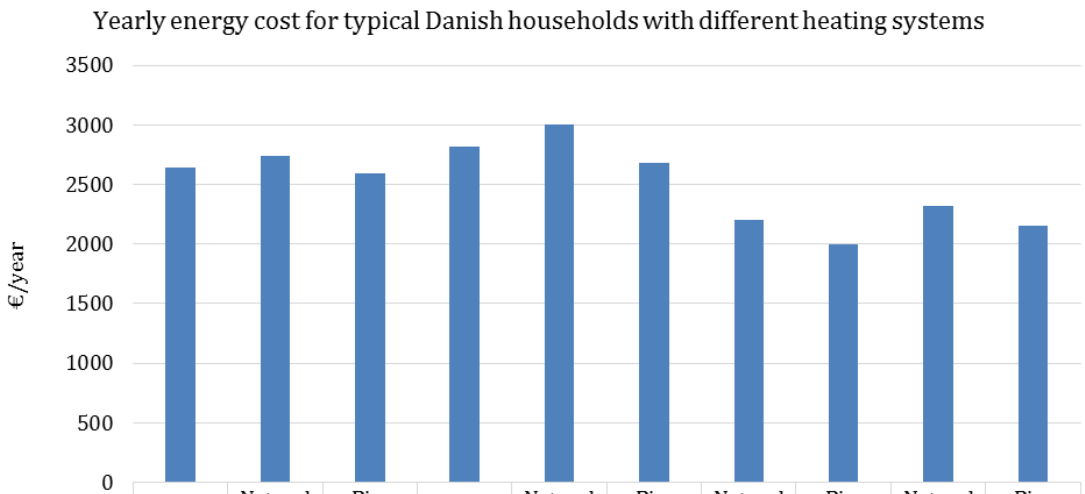


Figure 1. Yearly energy cost for typical household in Denmark.

At a closer look at the energy costs with traditional heating systems figures for district heating and boilers are very similar, the former resulting slightly less expensive.

The extra electricity consumption due to heat pumps providing all the heat demand raises grid power purchase from 4 MWh_{el}/y to 9.6 MWh_{el}/y. Arguably, this aspect may have pros and cons. On one hand, the flexibility and magnitude of such additional power demand can be exploited to enhance grid balance. However, suppliers have to provide suitable options to do so (eventually even in an aggregated fashion) which to date are very limited. On the other hand, such additional consumption may require large investments for upgrading the grid that are eventually paid by the customers.

Not surprisingly, optimal sizing and operation strategy of the fuel cell is key when it comes to minimize operating costs. Results confirm that micro CHPs are more suitable when used to meet local power demand and sized accordingly, as also reported in (ref 35 in the report). If larger natural gas units are sized to cover the heat demand (which in this case study is more than four times that of electricity), power sellback revenues do not offset the higher fuel consumption due to the lower thermal efficiency of the CHP vs. boilers.

In the modelled cogenerative fuel cell, one unit of electrical energy requires approximately four units of thermal energy from the gaseous fuel. Concurrently, the cogenerative heat displaces just less than two units of gaseous fuel (assuming a heat ratio of 1.5 and a boiler efficiency of 80%). Considering that fuel is purchased at the retail price which is above 70 €/MWh_{th}, ***hence, electricity is produced with an energy cost higher than 140 €/MWh_{el}***. This value is close to that of the sellback price (145 €/MWh, see section 1.4.5.2) of pure biogas based electricity. For natural gas, the power sellback price is comparable to that of the spot price (approximately 32 €/MWh_{el}) mining the economics of the application. Calculations show that revenues from power sellback are limited to 210 €/year when using natural gas. However, thanks to the strong incentives in place, revenues rise up to 1014 €/year when running on bio-methane (which is five times higher than on natural gas) as shown in Table 1.

Table 1. Heat and power consumption figures for typical Danish households with fuel cells in 2014.

	Fuel cell (with reformer) heat-led no boiler backup Case - A		**Fuel cell (with reformer) electricity-led boiler backup Case -B**		**Fuel cell (with reformer) part. electricity-led boiler backup Case - C**		
Fuel	**Natural gas**	**Bio-methane**	**Natural gas**	**Bio-methane**	**Natural gas**	**Bio-methane**	
Heat/power ratio	1.5	1.5	1.5	1.5	1.5	1.5	
Fuel price	76	69	76	69	76	69	[€/MWh]
Grid electricity price	300	300	300	300	300	300	[€/MWh]
Sellback electricity price	30	145	30	30	30	30	[€/MWh]

	Fuel cell (with reformer) heat-led no boiler backup Case - A		Fuel cell (with reformer) electricity-led boiler backup Case -B		Fuel cell (with reformer) part. electricity-led boiler backup Case - C		
Fuel	**Natural gas**	**Bio-methane**	**Natural gas**	**Bio-methane**	**Natural gas**	**Bio-methane**	
Heat consumption	16.2	16.2	16.2	16.2	16.2	16.2	[MWh/y]
Electricity consumption	4	4	4	4	4	4	[MWh/y]
Boiler efficiency	0.8	0.8	0.8	0.8	0.8	0.8	
CHP electrical efficiency [1]	0.26	0.26	0.26	0.26	0.26	0.26	
CHP thermal efficiency	0.39	0.39	0.39	0.39	0.39	0.39	
Total CHP efficiency	0.65	0.65	0.65	0.65	0.65	0.65	
Heat generation	16.2	16.2	2.7	2.7	1.3	1.33	[MWh/y]
Grid electricity consumption	[0.2	0.2	0.0	0.0	2.0	2.0	[MWh/y]
Electricity generation	10.8	10.8	4.0	4.0	2.0	2.0	[MWh/y]
Sellback electricity	7	7	0	0	0	0.0	[MWh/y]
CHP fuel consumption	41.5	41.5	15.4	15.4	7.7	7.7	[MWh/y]
Boiler fuel consumption	0.0	0.0	13.5	13.5	14.9	14.9	[MWh/y]
Total fuel consumption	41.5	41.5	28.9	28.9	22.6	22.6	[MWh/y]
Fuel cost	3157	2866	2198	1995	1714	1557	[€/y]
Grid elec-	58	58	0	0	600	600	[€/y]

[1] *Includes reformer (75%) and inverter (95%) efficiencies in addition to the system electrical efficiency of 36% (HHV based).*

	Fuel cell (with reformer) heat-led no boiler backup Case - A		Fuel cell (with reformer) electricity-led boiler backup Case -B		Fuel cell (with reformer) part. electricity-led boiler backup Case - C		
Fuel	**Natural gas**	**Bio-methane**	**Natural gas**	**Bio-methane**	**Natural gas**	**Bio-methane**	
tricity cost							
Electricity sellback (cost)	-210	-1014	0	0	0	0	[€/y]
Total energy cost	3005	1910	2198	1995	2314	2157	[€/y]

In in case A, a small amount of grid power consumption seldom occurs when the auto production is not sufficient to cover the instant demand (typically during summer when the heat demand is minimal, as shown in Figure 2.

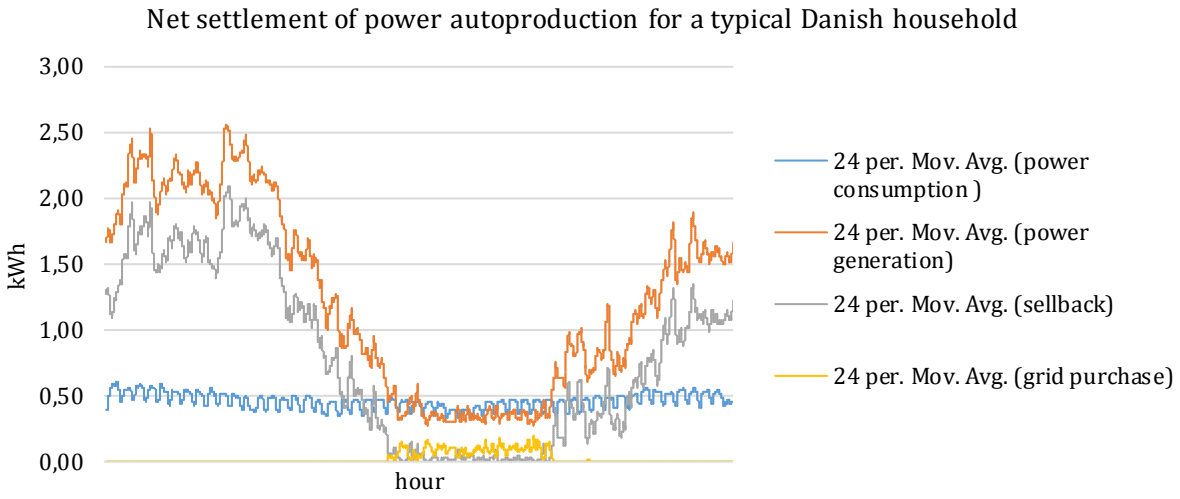


Figure 2. Net settlement of power auto production from a fuel cell for a typical Danish household during one year (heat- led operation, Case A). The power generation profile is determined by the heat demand which is met at any given time by the fuel cell (no heat storage is ideally required). During winter and middle seasons, auto generation exceed the power demand and electricity is sellback to the grid. During the summer season, a small fraction of the electricity demand has to be withdrawn from the main grid.

Discussion

This case study assesses the economics of using cogenerative fuel cells in typical Danish households. The total energy cost for covering yearly electricity and heat

demands was calculated for three different fuel cell sizes and compared to that of more traditional heating systems, namely district heating connection, gas boilers and heat pumps.

It was found that fuel cells can potentially reduce the energy cost from 2700 €/year to about 2000 €/year (by reforming bio-methane) or 2200 €/year (by reforming natural gas) when compared to traditional heating systems. However, in order to achieve such performance, the CHP unit must be designed and operate to avoid power sellback to the grid, as in case B and case C. As for the fuel cell size in case B, it is noticed that peak power consumption in Danish households are typically contractually limited at approximately 3 kW_{el}. Such size will definitely ensure that the system can meet the demand at any given time. In order to reduce installation costs of fuel cell systems, it is suggested that coupling the CHP with an electrical battery that would cover peaks that occur very seldom allowing for significantly smaller fuel cells.

It must be noticed that when the amount of grid power purchased is very low (case A) or even zero (case B), results may be bias. This is because utility bills were modelled proportionally to the average retail price and total consumption without fix costs. Whilst this method is effective for typical consumption values, it tends to underestimate the cost as it decreases. The extreme case is case B, where no grid electricity is consumed at all and according to Table 1 the grid purchase cost is zero. However, plant owners may be willing to keep the electrical grid connection (unless backup generation is available) that comes at a minimum fix cost.

Arguably, the study could have modelled electrical storage and advanced operation techniques for achieving higher revenues from power sellback in case A. However, it was found that the electricity production cost is too high to be matched by sellback payments most of the times.

Results also show that, when available, bio-methane is an attractive option thanks to favourable taxation schemes. Hence, it is suggested to further investigate on the biogas availability for residential reforming that may have a significant potential especially in East Denmark where numerous pig farms can provide a large amount of biomass to be processed.

Although our analysis shows that fuel cells may reduce total energy costs, it is advised to consider that payback periods may be very high for complete fuel cell systems that include reformer and power electronics (see Table 2). More details on simulation can be found in the report.

Table 2. Payback Period (PB) for increasing fuel cell system investments costs. Calculations assume 500 €/year savings as calculated.

PB (years)	5	10	15	20
Investment (€)	2500	5000	7500	10000

1.4.1.4 Specifications relevant for fuel cell operation in the electricity market

In this section relevant information is provided on key parameters for fuel cells operating in the electricity market. It includes aspects of lifetime, cycling, degradation and recommended reference operation profiles.

First, the lifetime and performance degradation patterns are described while operating a fuel cell under such flexible conditions. To do so, a testing strategy and a method to calculate degradation coefficients are presented that are meaningful when assessing the economics of fuel cell systems that highly depend on the stack installation and replacement costs. This procedure is applied to available experimental data from the HT-PEM membrane considered within the SmartMEA project. Finally, a suitable time series is provided to be used for future tests. This data was obtained by processing results from the simulations described in the report.

1.4.1.5 Lifetime and performance degradation

Simulating the operation of fuel cell systems over long periods of time may often require considering performance degradation and expected lifetime. Models are available with various degrees of complexity that describe the physical and chemical mechanisms governing degradation patterns. However, including such models in simulation tools for the operation of electrical systems as in the context of our study may not be practical. A more suitable model for modelling the health of a fuel cell over its operating period is to experimentally determine specific parameters that describe the degradation effect of a specific operation event. More specifically, we are interested in determining:

- The degradation effect on lifetime and efficiency resulting from a certain period of continuous operation at constant power output.
- The degradation effect on lifetime and efficiency resulting from a start-up event.

Such degradation patterns formulation can easily be included in simulation algorithms for electrical systems. The lifetime deterioration cost and performance reduction can become a decision variable of mixed integer linear programming, MILP optimization or, alternatively, they can be treated as parameters updated ex-post after each iteration.

Available literature provides insights on different metrics associated with the degradation of single MEAs, stacks and fuel cell systems. Although the amount of tests available in literature is extensive, the assessment of fuel cells degradation patterns is a complex task due to the lack of standard protocols. This study contributes to extrapolate degradation coefficients from experimental data available in literature.

1.4.1.6 Degradation coefficients for the Smart-MEA HT-PEM

Based on data from (ref 31 in the report), the following was derived; the V_{OC} degradation coefficients of 8.1 µV/h and 67.4 µV/h resulting from one hour of fuel cell operation at 0.03 A/cm^2 and at 0.25 A/cm^2 respectively. Also it was calculated that a single start-up procedure contributes to a 115 µV of V_{OC}^0 reduction see Figure 3.

In Figure 3, calculated degradation coefficients were provided of the voltage at the same two current levels due to start-ups and continuous operation.

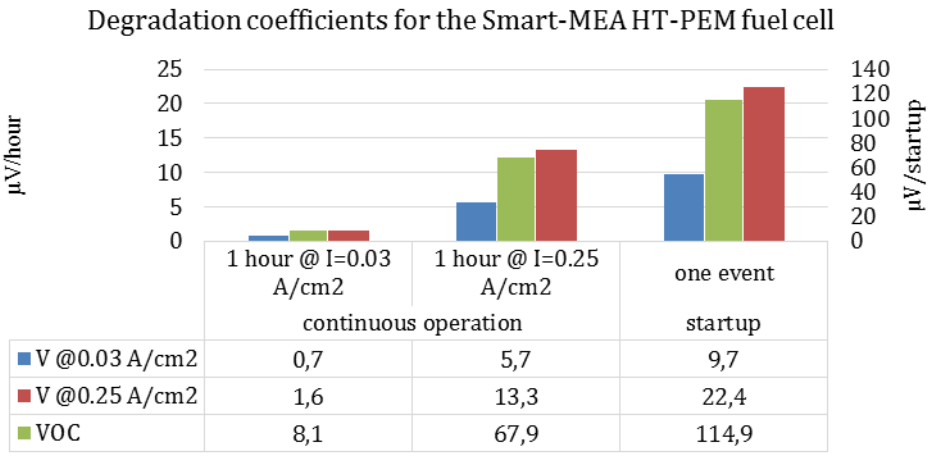


Figure 3. Calculated voltage degradation rates due to continuous operation at two current levels and start-up events for the cycling experiment of the Smart-MEA HT-PEM fuel cell in [ref 31 in the report]. Due to long periods of operation at open circuit voltage condition associated with very high degradation, given values are likely overestimated, as pointed out by the authors of [ref 31 in the report].

Degradation coefficients expressed as in Figure 3 are more suitable for modelling the expected lifetime and performance degradation patterns for grid applications that typically require numerous start-up and part-load operation of dispatchable units. For example, estimated end of life is reached after approximately 3100 hours of continuous operation at 0.25A/cm^2 or 26,000 hours at 0.03A/cm^2. Cycling the cell with one start-up per hour would shorten the lifetime to 1200 hours if operated at 0.25A/cm^2 whilst 1700 hours can be expected if operated at the lowest current output.

In absence of data for other current density levels the maximum current density is 0.40A/cm^2, a linear interpolation of the operating degradation rates was used to determine the continuous operation V_{OC} degradation coefficient λ_{OC} in μV/hour as a function of the current in p.u. $I_{pu}(t)$:

$$\lambda_{OC}(t) = 107.9\ I_{pu}(t)$$

The open voltage circuit at a given time can be formulated as a function of the number Kt of start-up events occurred since the fuel cell installation and of the operating degradation coefficient:

$$V_{OC}(t) = V_{OC}^{0} \cdot [1 - (K(t) \cdot \mu_{OC}) - (u_{on}(t) \int_0^t (\lambda_{OC} \cdot t \cdot \delta \cdot t))]$$

Where μ_{OC}=115 μV/start-up is the startup V_{OC} degradation coefficient and u_{on} is a binary switch that takes zero when the current is zero.

Voltage degradation effects are more noticeable when the unit is operating at higher power rates 6.7% at 0.25 A/m^2 vs. 2.4% at 0.03 A/m^2. By assuming that the efficiency reduction is proportional to that of the voltage and varies linearly across the operating range, the end of life stack efficiency η_{stack}^{EoL} can be determined at a given current I_{pu}:

$$\eta_{stack}^{EoL}(I_{pu}) = \eta_{stack}^{BoL}(I_{pu}) \cdot [1-(0.078\ I_{pu}+0.018)]$$

where η_{stack}^{BoL} is efficiency for a newly installed unit. This formula allows determining the end-of-life efficiency curve from that of a newly installed one with efficiency η_{stack}^{BoL} as shown in Figure 4 where values from a LT-PEM was used due to lack of

efficiency data for the case study [ref 31 in the report]. At any given time t and current density I_{pu}, the stack efficiency can be calculated from η_{stack}^{EoL} and $V_{OC}(t)$:

$$\eta_{stack}(t, I_{pu}) = \eta_{stack}^{EoL} \cdot (I_{pu}) \cdot (V_{OC}(t))/(V_{OC}^{0})$$

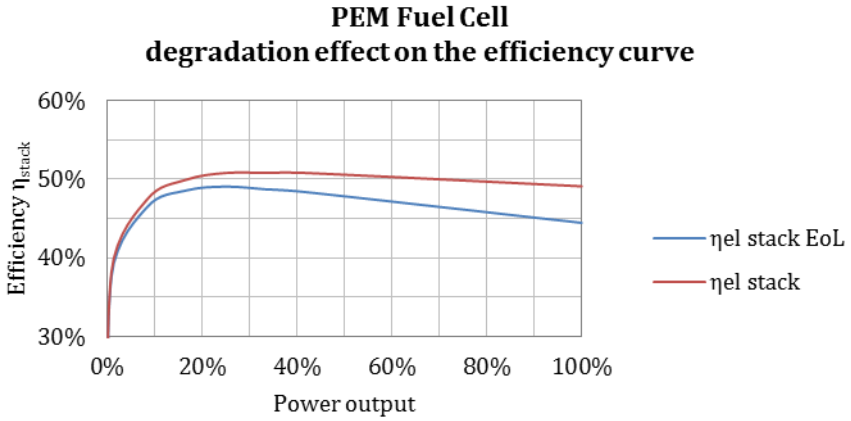


Figure 4. Degradation effect on the stack efficiency curve of the PEM fuel cell from Figure 13 calculated with the calculated coefficients from [ref 31 in the report]. At its end-of-life EoL, efficiency reduction is more noticeable at higher power rates.

1.4.1.7 Degradation test protocol recommendation

In this section, it was the goal to determine degradation coefficients suitable for electrical system simulations from experimental data available in literature. It was found that only one experiment [ref 31 in the report] was useful for the purpose thanks to the following characteristics:

- Well defined operating cycles in terms of duration and current density
- Presence of start-up events within cycles
- Voltage measurements at multiple operating points repeated every cycle
- Long testing period

The following recommendations are provided for testing a fuel cell performance with regards to power grid applications and flexible operation in general. This procedure was used to determine degradation coefficients reported in Figure 16 and used to derive ⟦ λ⟧_OC and⟦ η⟧_stack^EoL in the equations above in this section. It is advised to perform one cycling test based on the typical operating cycle required by the specific application considered and a number of continuous operations at constant load.

Cycling test

K repetitive cycles are considered indexed with k=1,…,K. During the cycle, consider n intervals indexed with i=1,…,n and j=1,…,n of duration Δt_i during which a constant current density $I_{pu,I} > 0$ is maintained. During cycle k, the cell voltage V_ik is conveniently measured for each interval Δt_i and it is used to calculate the mean cycle voltage degradation rate in μV/cycle occurred throughout the test when measured at the current density I_i:

ΔV_i^cycle=($\sum_{(k=2)}^{K}$·[V_i(k-1)-V_ik])/K

If during the cycle the unit is switched off for simplicity one turndown is assumed, and also that the open circuit voltage V_oc is measured and calculated by mean value:

$\Delta V_{OC}^{cycle} = (\sum_{(k=2)}^{K} \cdot [V_{OC}(k-1)-V_{OC}k])/K$

Because operating the fuel cell at open circuit voltage conditions creates high cathodic potentials resulting in very high degradation rates, it is advised to avoid such state for longer periods, as pointed out in [ref 31 in the report]. Hence, the effect of this operating condition is neglected.

Obviously, throughout a cycle the cell degrades during each time interval. It is assumed that the voltage reduction during one interval is proportional to its duration:

$\Delta V_i^{cycle} \propto \sum_{(j=1)}^{n} (\alpha_{ij}\Delta t_j)$

$\Delta V_{OC}^{cycle} \propto \sum_{(j=1)}^{n} (\alpha_{OCj}\Delta t_j)$

Where α_{ij} and α_{ij} are unknown coefficients in μV/hour. A lump voltage degradation coefficient μ_i and μ_{OC} in μV/startup are associated with the start-up event that follows the off period and include it in the previous proportions

$\Delta V_i^{cycle} = \sum_{(j=1)}^{n} (\alpha_{ij}\ \Delta t_j) + \mu_i$

$\Delta V_{OC}^{cycle} = \sum_{(j=1)}^{n} (\alpha_{OCj}\ \Delta t_j) + \mu_{OC}$

Similarly to α, also μ is unknown at this stage.

Continuous operation test

In order to determine the unknown parameters α_{ii}, a total of n other experiments are carried out. During each of these experiments, the current density level will be kept constant at $I_{pu,i}$ and the voltage V_i will be continuously logged. From these measurements , the average degradation rate α_{ii} is calculated.

To calculate the remaining unknowns, it is assumed that

$\alpha_{ij} = \alpha_{ii}\ (I_{pu,j})/(I_{pu,i})$

and that

$\alpha_{OCj} = \alpha_{jj}\ (\Delta V_{OC}^{cycle})/(\Delta V_i^{cycle})$

Finally, the i+1 start-up degradation coefficients μ is calculated by solving the resulting system of i+1 linear equations.

Reference operating profiles

A normalized hourly fuel cell power operation throughout the year is plotted in Figure 5 based on the report. The overall trend can be observed in Figure 6 for a typical day. However, a high degree of variability dominates the operating patterns. In particular, it is noticed that the fuel cell is used more intensively during late summer and autumn whilst in late winter and early spring is operated at lower rates. The morning and evening peaks as well as the low power rate during night are easily recognized.

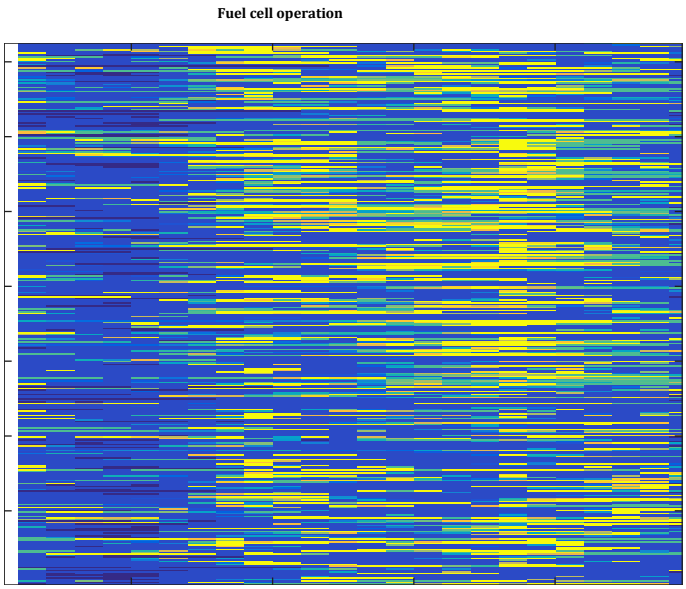


Figure 5. Normalized fuel cell hourly operation throughout one year. Data are from the reference scenario (REF) simulated in the report.

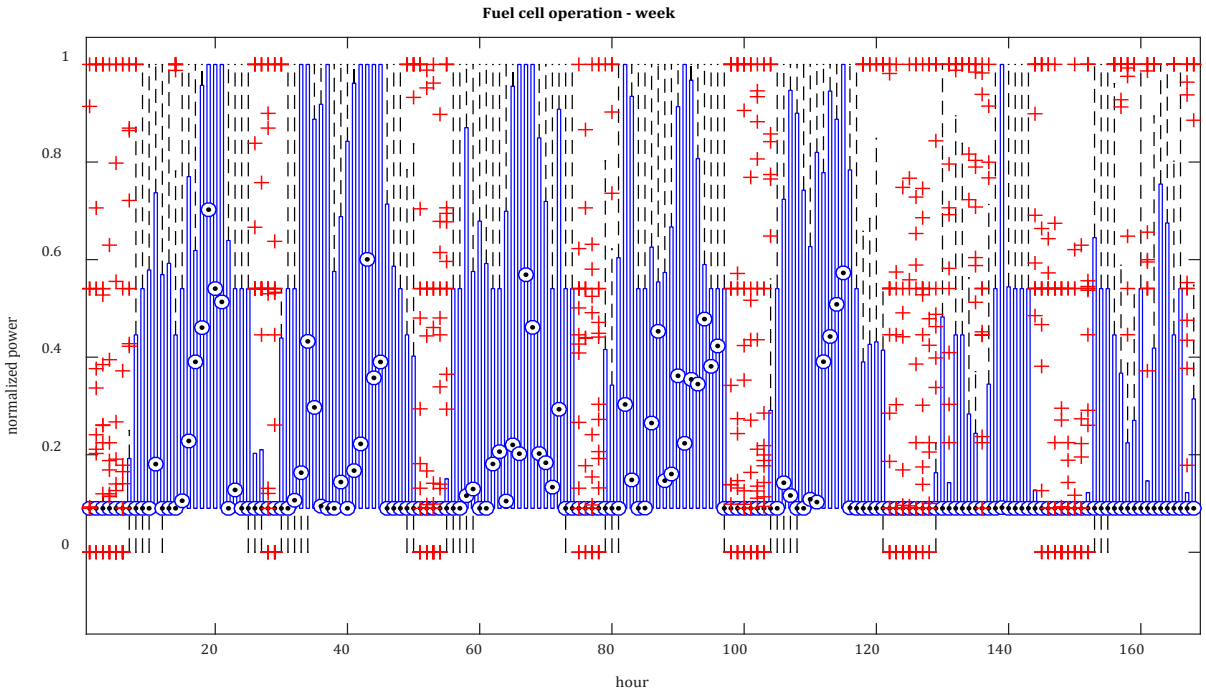


Figure 6. Statistics for weekly fuel cell operation. Data are from the reference scenario (REF) simulated in section 2.4 of the report. On each box, the central mark is the median, the edges of the box are the 25th and 75th percentiles, the whiskers extend to the most extreme data points not considered outliers, and outliers are plotted individually.

Statistics for the weekly fuel cell operation is plotted in Figure 6 where the two-peak daily pattern is observed, especially in the percentile boxes. It shows more clearly that variability of the operating schedules. Power fluctuations can be very ample throughout the whole operating range above the turndown point (it is never switched off in the late afternoon and early evening) during the day whilst at night the unit is more likely operating at its minimum power and at times turned off.

The cumulative distribution of the fuel cell power is shown in Figure 7. The stepwise profile indicates four power levels that the unit is keener to work at. During 3.9 % of the time the fuel cell is switched off and does not produce power. Most of the time (57 %) the unit is kept working at its turndown ratio. The third step represents an intermediate power output occurring when the fuel cell is directly fed by the reformer and electrolyzer and not by the storage. This situation happens about 7 % of the time. Finally, fuel cell output is maximal during approximately 13 % of the time.

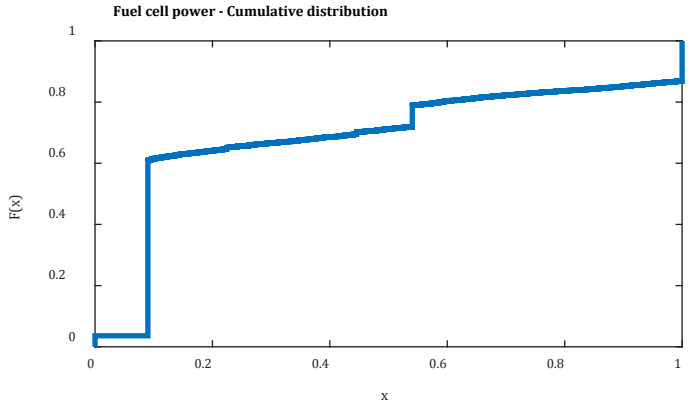


Figure 7. Cumulative distribution F(x) of the (normalized) fuel cell power x. Data are from the reference scenario (REF) simulated in the report.

1.4.2 WP 2. Next generation electrodes

(Lead: DTU Energy)

1.4.2.1 Effect of gasdiffusion layers on phosphoric acid retantion

The effect of the material that the electrode made from, i.e. the porous gas diffusion layer (GDL) that the catalyst layer is applied on, was studies in the PhD project at DTU Energy. The aim was to see if phosphoric acid loss and cell durability was affected by the properties of the GDL. One may expect that this electrode backing plays a minor role in acid retention since the acid is in the membrane and in the catalyst layer. The GDL is wet proofed, i.e. treated with Teflon to become hydrophobic and phosphoric acid should not easily wet the GDL.

Four different materials for the use as GDL and electrode backing from the company Freudenberg GmbH were tested systematically. The materials were H23C2 (C2), H23C4 (C4), H24C3 (C3) and H24C5 (C5). Short names in brackets. C2 is the ma-

terial recommended for HT-PEMFC by Freudenberg and the material most often used.

Passive acid uptake measurements showed that the GDLs C2 and C4 sucked up much more acid that the GDLs C3 and C5. See Figure 8.

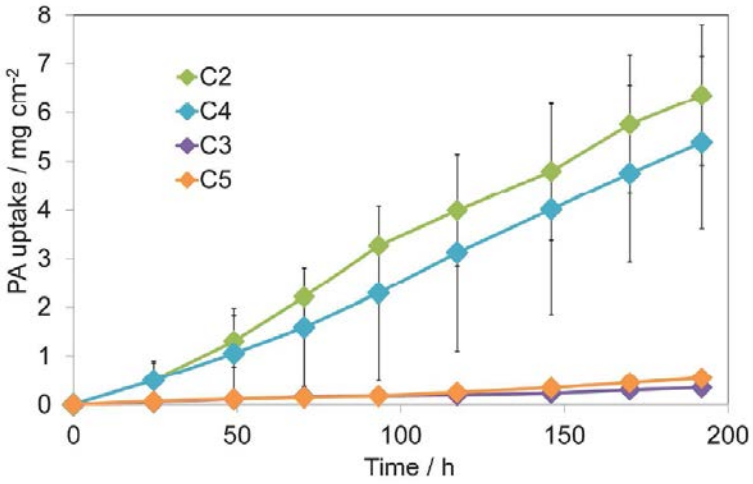


Figure 8. Acid uptake for the GDLs C2, C3, C4, and C5. The error bars represent the total span of 9 measurements for each data point.

Mapping of the phosphoric acid by energy dispersive x-ray spectroscopy (EDS) of cross sections of the tested samples are shown in Figure 9. It is evident that the fine microporous layer (left most in the figures) had taken up the most acid, but the coarse fibrous backing layer actually contains some acid too. It is also evident that the materials C2 and C4 contained the most acid in the micro-porous layer as well as in the fibrous layer. This is if perfect agreement with the results in Figure 8.

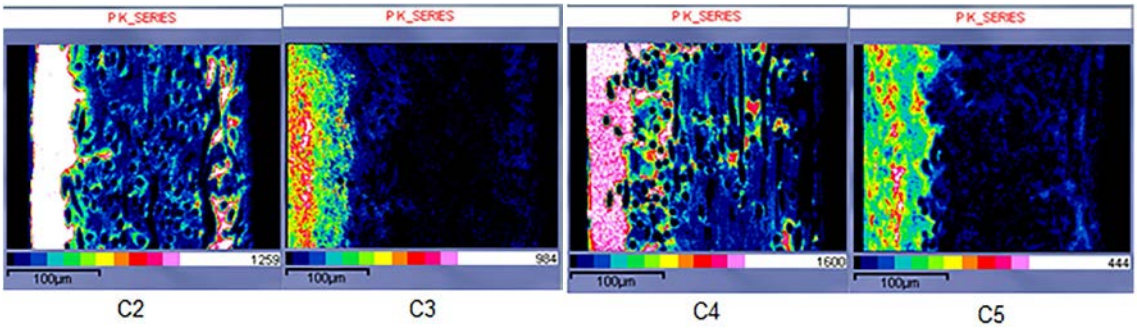


Figure 9. EDS mapping of Phosphorus distribution in the GDL materials after acid uptake over 200 h. The microporous layer in contact with the acid is located in the left side of the images. The remaining major parts are the fibrous layers. The colour coding indicates count density and highest values are given in the white fields.

Fuel cell durability tests were performed systematically with different combinations of GDLs in the electrodes. Figure 10 shows selected cells which all keep C2 for the anode (the hydrogen electrode) and varied GDLs for the cathode (the oxygen electrode). In the course of the testing period the cells with C3 and C5 show more stable performance and less degradation that the cells with C2 and C4. Especially the cell with C2 cathode degrades faster. Corresponding acid collection measurements

from the cathode exit indicate acid losses in nice accordance with the performance decay.

The conclusion drawn was that degradation in the long term tests is to a large extent a consequence of acid loss. This loss is more pronounced in the cell with the recommended GDL. The mechanism is most likely that phosphoric acid diffuses most easily into the GDL in this case and that the large surface it thus spreads over eases evaporation of the acid which is then carries out of the cells with the gas flow.

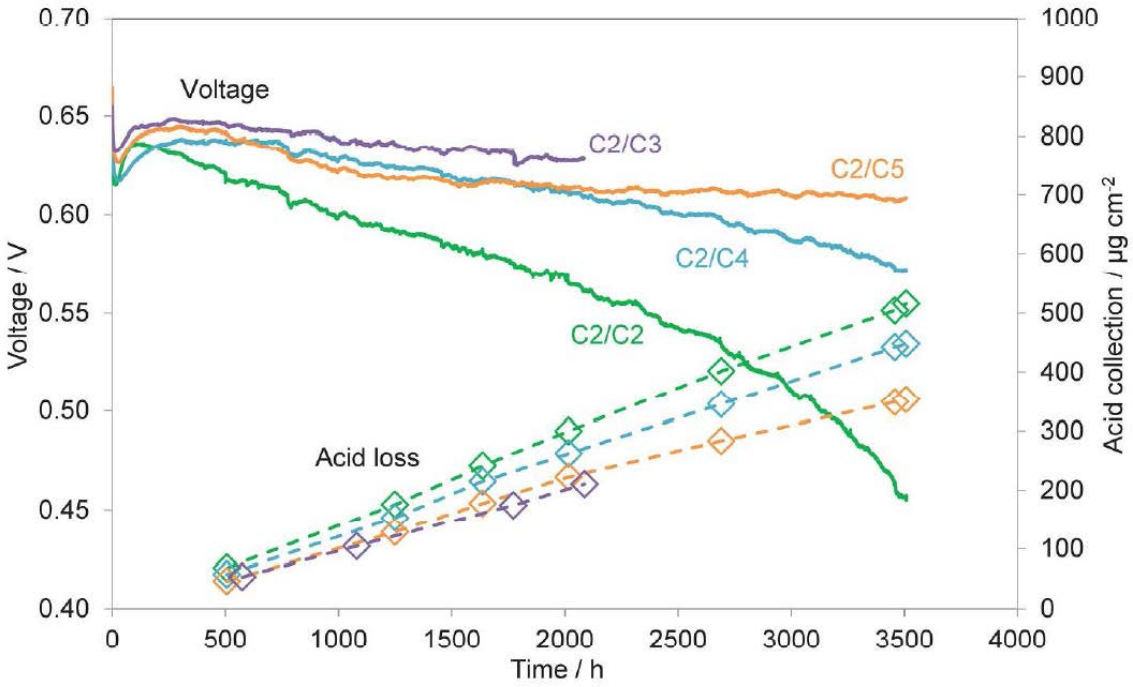


Figure 10. Durability of cells with varied GDL on the cathode side (solid lines) and cumulative acid collected at the fuel cells exhaust (dotted line). The cells were operated at 180 °C, 200 mA cm^{-2}, λ_{H2} = 1.5 and λ_{air} = 2. The cell identification is ordered as anode/cathode GDL.

The amount of acid collected at the cathode exhaust was only a few percent of the total acid in the system, so a major part of the lost acid was probably halted on the way out by precipitation different places in the cell. Nevertheless, A reasonable correlation between the acid collection rate and the increase of the series resistance (ohmic resistance – mostly from the membrane) was seen (Figure 11).

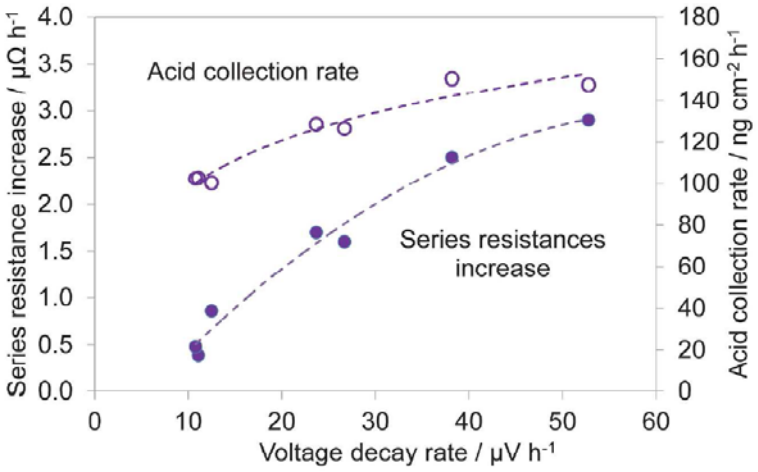


Figure 11. Acid collection rate and series resistance increase rate as a function of voltage decay rate.

[The study of the effect on acid loss of the GDL is important since it appears that the material recommended and most often used, H23C2 (or just C2), is the one with the poorest acid retention capabilities. The finding were presented at the firth International Carisma Conference in Newcastle [2] and published in Fuel Cells [3]

1.4.2.2 Electrode manufacture and platinum loading

DPS have invested a significant effort in both electrode- and process development:

- Implementation of new alloy catalyst (lower Pt loading and higher performance)
- Upscaling of manufacturing process (lower cost and higher capacity)

The activities within SmartMEA have contributed significantly to these activities.

Reduction of platinum content

Platinum (Pt) is the active catalyst in the fuel cell but also a precious metal with a high price. DPS has been part in several research projects investigating the effect on performance and lifetime by using a different catalyst. Here it has been concluded that using a commercially available PtCo/C catalyst gives the same or slightly higher performance with use of 1/3 less Pt. The implementation of the use of this catalyst has been combined with the implementation of a new production technique (tape casting) which is more suited for high volume production and much cheaper. The current product is based on electrodes manufactured through a spraying process applying up to 100 layers of catalyst on the carbon substrate. The new tape casting process applies the catalyst in one layer / one pass enabling a much higher production capacity and reduced price.

[2] A. Kannan, Q. Li, L.N. Cleemann, J.O. Jensen. Durability of HTPEM fuel cells with different electrode supports. Poster presented at the 4th international CARISMA Conference, Newcastle, United Kingdom, (2017)

[3] A. Kannan, Q. Li, L. N. Cleemann, J. O. Jensen. Acid distribution and durability of HTPEM fuel cells with different electrode supports. Accepted for publication in Fuel Cells (2017)

The performance of MEAs with the tape casted electrodes has been tested substantially with hydrogen and wet reformate.

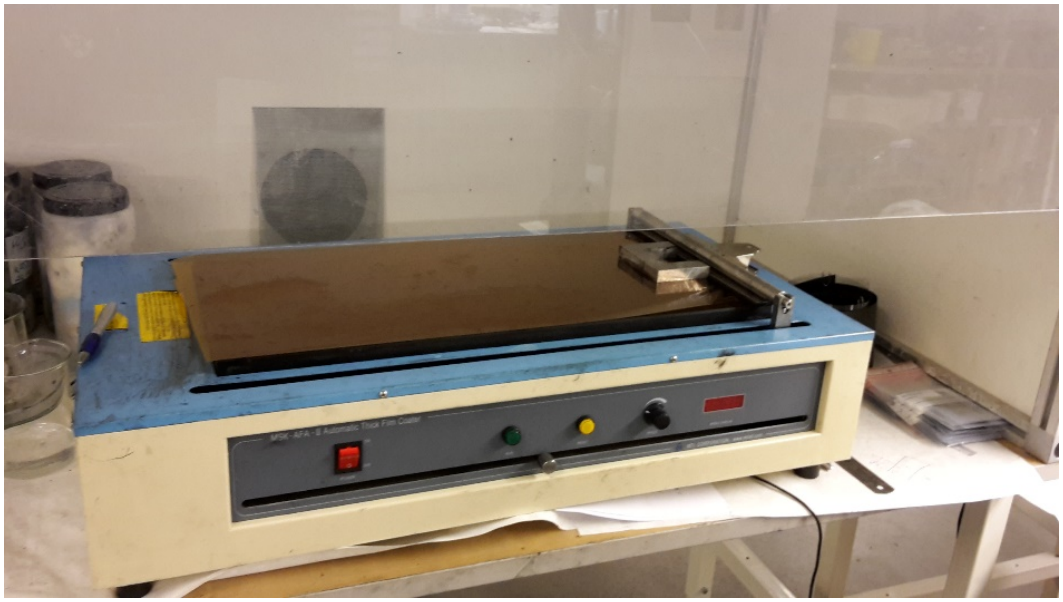


Figure 12. Table top tape casting equipment for development of new electrode production method.

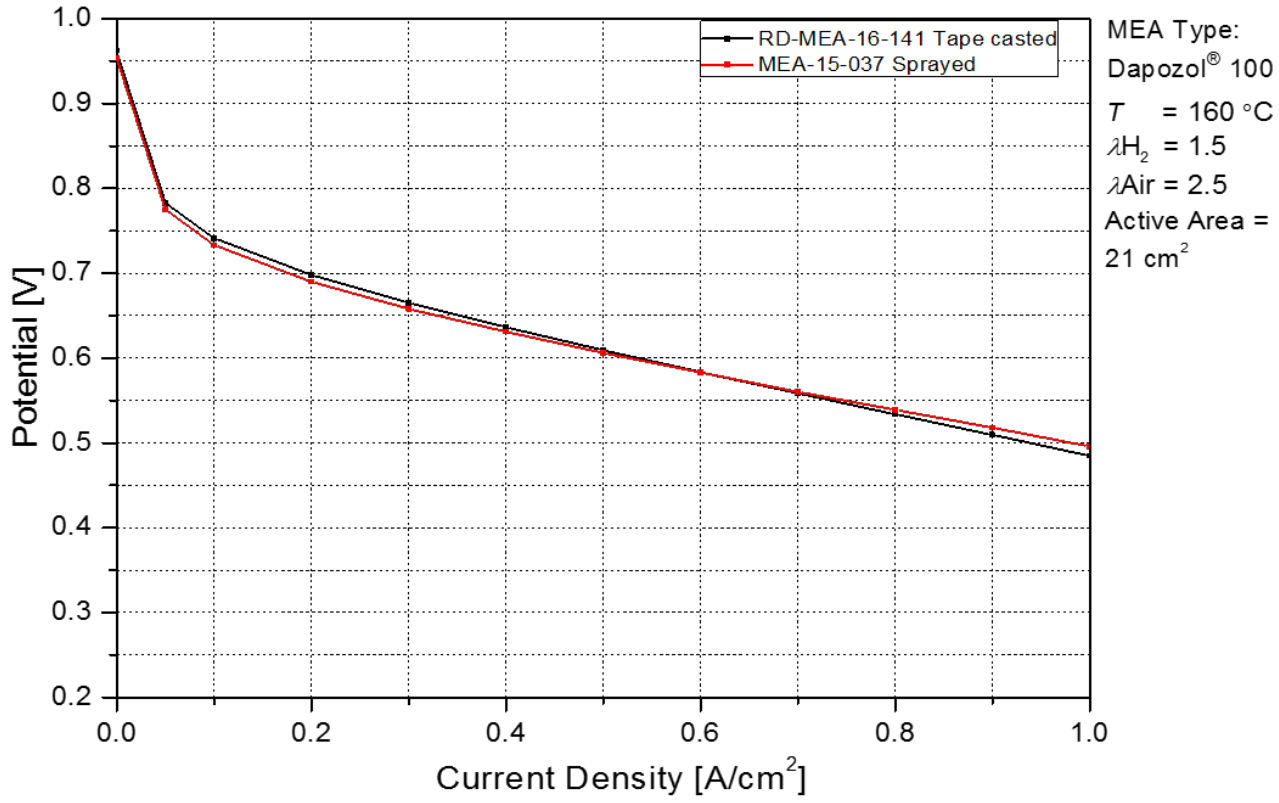


Figure 13. Comparison of sprayed electrodes based on Pt/C catalyst (A/C: 1.5 / 1.5 mg/cm^2) vs. tape cast electrodes based on PtCo/C catalyst (A/C: 1 / 1 mg/cm^2).

The new electrodes based on the PtCo/C catalyst have the same performance as the electrodes based on the existing Pt/C catalyst despite having a 40 % reduction in Pt loading.

1.4.2.3 Electrospinning of electrode structures

Electrospinning has been widely used to construct 3-dimensional porous structures of fuel cell electrodes. In this WP, DTU Energy carried out electrospinning activities to design electrospun nanofiber gas diffusion layers (GDL).

Upgrading of electrospinning instrument

Safety evaluation of electrospinning process was conducted during SmartMEA project. To ensure no solvent and nanoparticles can escaped from electrospinning instrument, a modification to the existing system was carried out. The design and construction was done by electrospinning instrument vendor Linari Engineering s.r.l.

As shown in Figure 14, an internal chamber, of reduced dimensions compared to the existing window, was added to allow the suction of gas leaks due to the cavity that is created. In the roof-top, the existing suction system with an automatic butterfly intake valve regulates the suction of the gases present in the chamber. Additional equipment to ensure the suction of leakages in the cavity: a perimetral suction channel. This system was constituted by tubes that joint on a C-shaped suction channel that aspire gases present in the space between the two chambers through dedicated holes in the roof-top. The internal chamber had an openable door (flap) for the operations requiring more freedom of movement, and two sliding windows for the operations carried out during the suction of the gas. A sensor verifies the correct closure of the flap prior to closing the outer window.

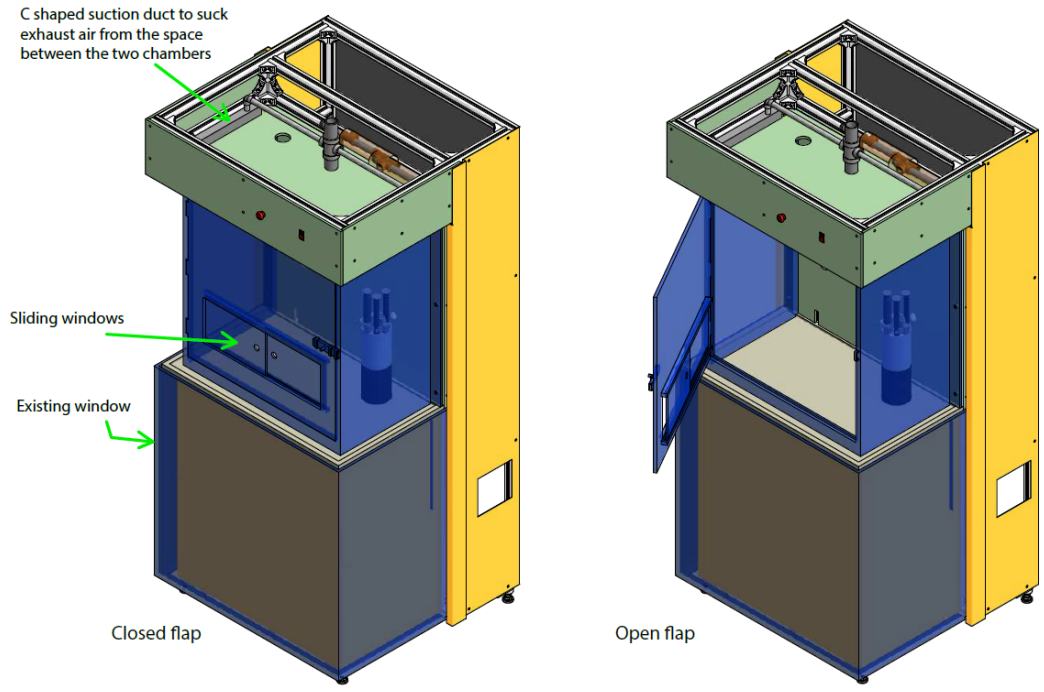


Figure 14. Schametic diagram of upgraded electrospinning instrument.

Electrospinning was then utilized to shape the resulting ink into nanofiber layer, as shown in Figure 15a. The diameter of PVP nanofibers is in the range of 200-400 nm (Figure 15b). To convert the PVP carrier into a carbon support, we carbonized the resulting nanofiber layer in N_2/H_2 at 800°C. The diameter of carbonized nanofibers is was reduced to ~50 nm.

a b

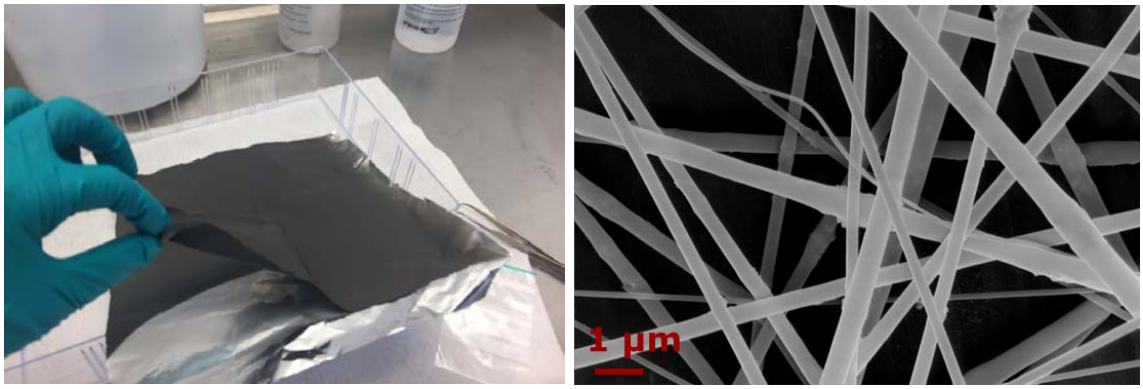


Figure 15. a) As-spun Pt-C/PVP nanofiber layer peeled off from Al foil; b) SEM image of the nanofibers.

Conventional carbon GDL is composed of carbon fibers that have average diameter of ~14 µm and pore size of more than 20 µm, as shown in Figure 16a. This imposes critical challenges to apply Nano sized catalysts on top of GDL, where the catalyst ink is trapped into micron-sized pores between carbon fibre. Nanofiber carbon GDL was made by electrospinning followed by carbonization. The resulting structure is 100 times finer as compared to commercial GDL, as shown in Figure 15b. Such structure can be applied as interface layer between conventional GDL and catalyst layer, to optimize catalyst distribution and enhance mass transport.

a. **b.**

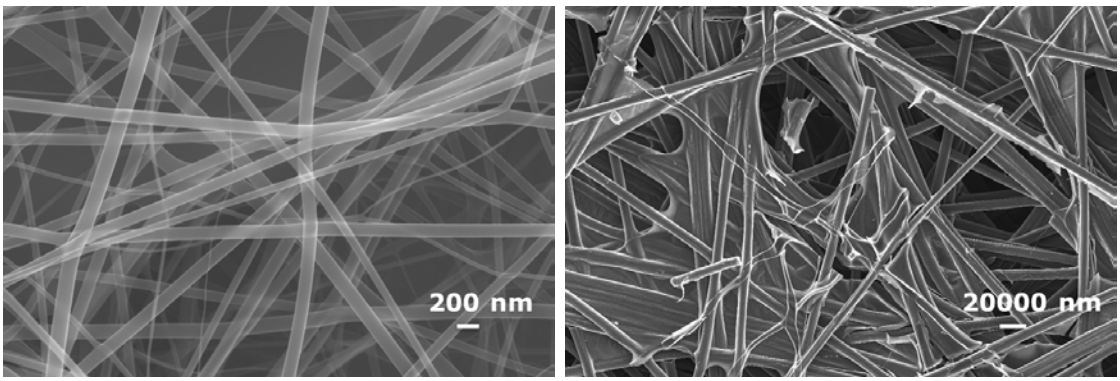


Figure 16. a) SEM image of electrospun carbon nanofibers; b) SEM image of commercial GDL.

1.4.2.4 Platinum catalyst thermal stability

It is well-known that platinum nanoparticles tend to grow by a range of different mechanisms. Since the class of fuel cells in the project is operated at elevated temperature it was decided to study thermally induced particle agglomeration. By means of in situ transmission electron microscopy (TEM), the thermal coarsening of Pt nanoparticle was monitored during the exposure to 6 mbar 20% H_2/Ar while ramping up the temperature to ca. 900 °C. Time-resolved image series directly reveal that separated 3 nm sized Pt nanoparticles in a hydrogen environment are stable when the temperature is lower than ca. 800 °C. Furthermore, the coarsening above ca. 800 °C is fully dominated by the particle migration and coalescence mechanism. Contrary to Pt nanoparticle coarsening in oxygen, the Pt coarsening in hydrogen was not mediated by an Ostwald ripening. For agglomerated Pt nanoparticles, the coalescence was observed already at the temperatures above ca. 200 °C. The temperature-dependency of particle sizes and the average migration distance is described and found to be consistent with simple early models for the migration and coalescence. Figure 17 presents an in situ TEM images series of the Pt/CB catalyst during temperature ramping at 10 °C/min from 50 °C to 960 °C in 6 mbar of 20% H_2/Ar. Details can be found in the paper published [4]

1.4.3 WP 3. Enforcement technologies

Lead: DTU Energy

1.4.3.1 Fibre enforcement

Work on fibre enforcement was planned in the project description. One way that has been initiate in the ForskEL project HotMEA, was electrospinning of PBI fibres and the idea was to crosslink the spun fibres to improve strength and subsequently to fill the voids with “simple PBI. This was not pursued. It was delayed due to a maternity leave and later priority was shifted in favour of another way to modify the membrane as reported in part 1.5.5.1 Durable membrane on page 31.

[4] Søren Bredmose Simonsen, Yan Wang, Jens Oluf Jensen, Wenjing Zhang. Nanotechnology, 28, 475710 (2017)

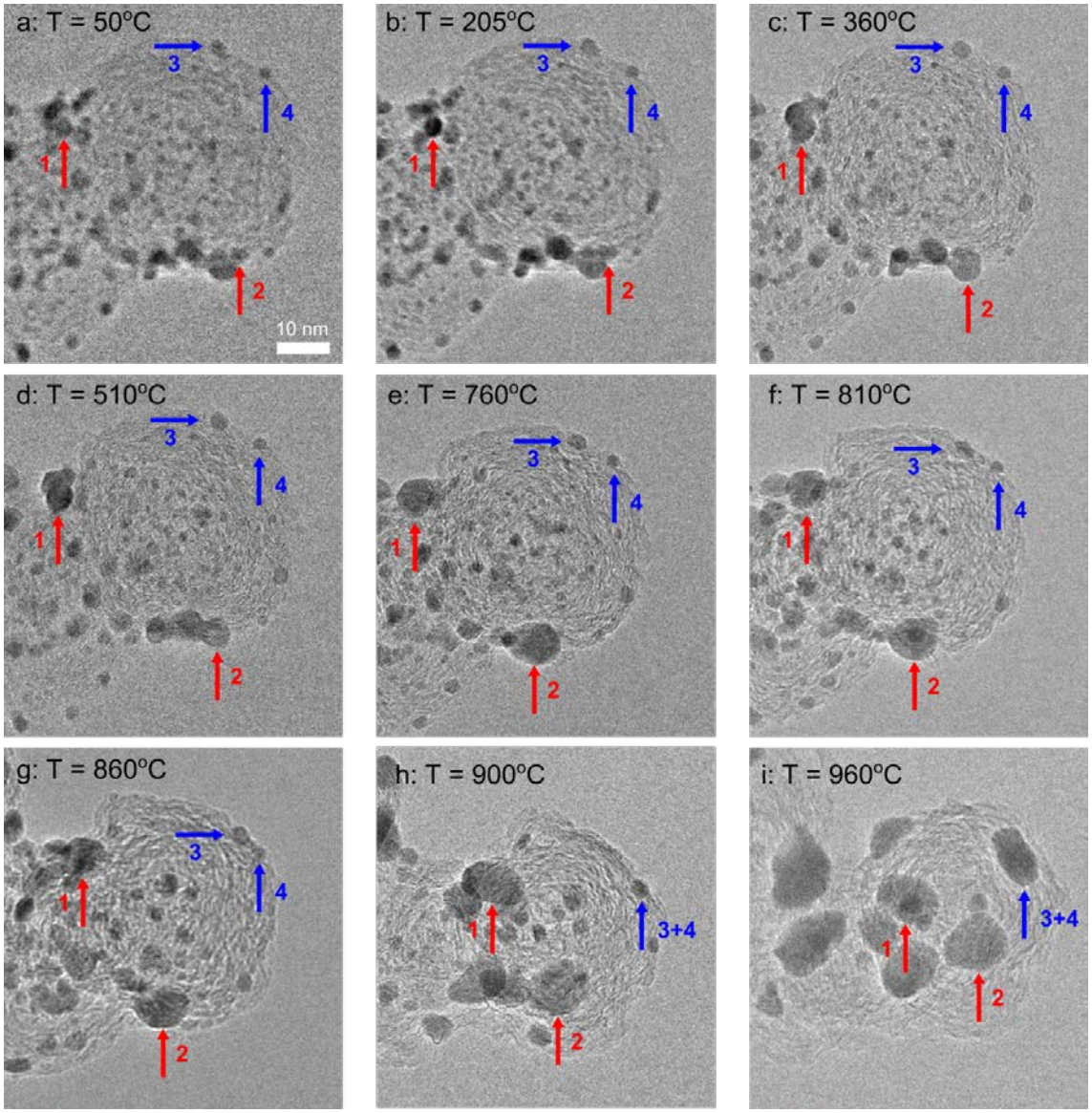


Figure 17. (a-i) In situ TEM images of Pt nanoparticles on a CB support during temperature ramping at 10 °C/min and exposure of 6 mbar 20 % H_2/Ar. Temperatures are indicated in the images. Examples of two types of Pt nanoparticles are indicated by arrows: agglomerated (red) and separated (blue).

1.4.3.2 Sub-gaskets and sealing

Sub-gasket is an enforcement technique that has been used prior to SmartMEA, but is still an important part of the enforcement strategy

A bonding process between the electrode and the sub-gasket have been developed in order to improve the mechanical integrity of the MEAs. Furthermore, the required gasket (needed for a leak tight stack) have been integrated on the sub-gasket (See Figure 18).

A stack consists of a high number of MEAs, sealing and bipolar plates placed on top of each other. For the stack to work properly it is important to ensure the correct compression and gas tightness of the stack. The correct placement of the different components on top of each other is therefore essential.

DPS has developed a method to place the gasket onto the MEA before cutting out the gas manifold holes. Subsequently the gas manifold holes are cut in the MEA and gasket at once. This has eliminated the difficult assembly step where the MEA with cut-outs and the sealing with the same cut-outs had to be placed correctly on top of each other.

Previously this operation was done by the customer but it was a very difficult and time consuming process. Now by moving this to the MEA manufacturer (DPS) it can be done at an earlier stage and as part of the MEA manufacturing. Moving this process to the manufacturer has also improved the final product (MEA + sealing) as it is much easier to adjust the MEA manufacturing QC requirements to the sealing specifications when handled by the same party.

This potentially also enables the replacement of the current sub-gasket with the gasket, thus further potential savings.

Figure 18. MEA with gasket attached and gas manifold holes cut out in both materials at the same time.

1.4.4 WP 4. MEA manufacturing

(Lead: DPS)

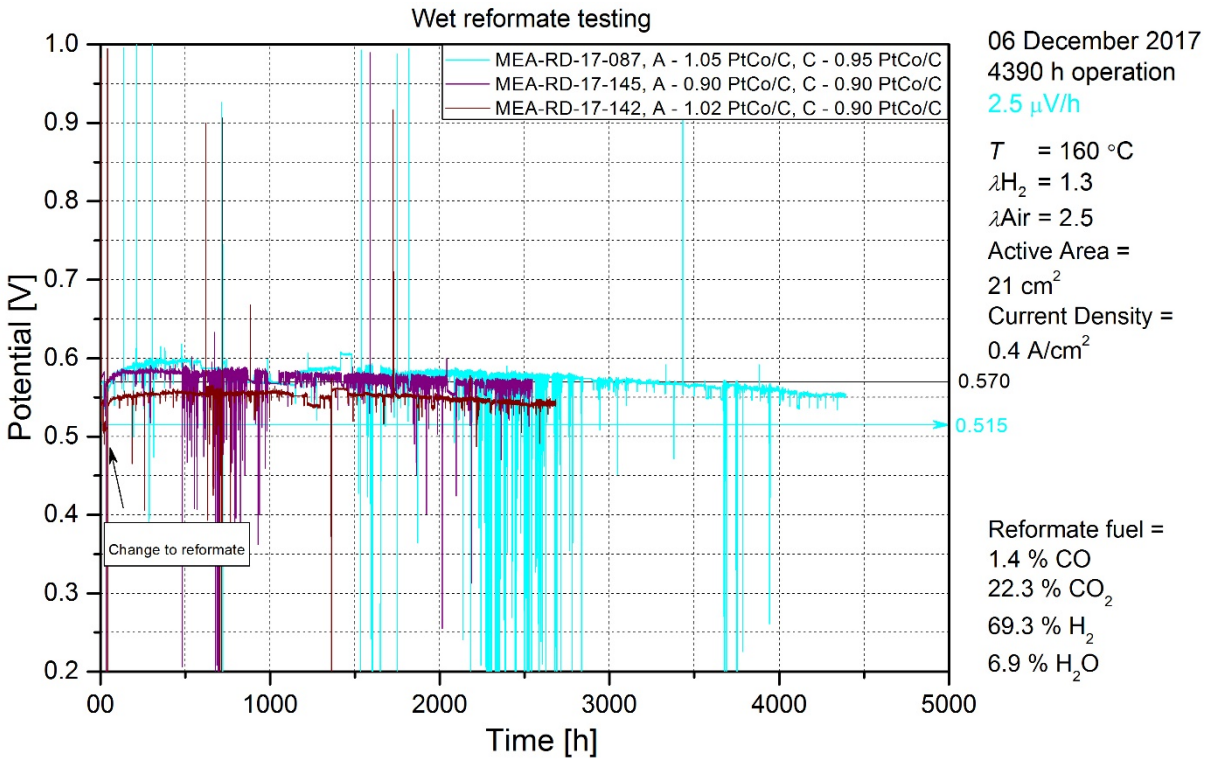
Tape casting was used for the initial ink formulation and process development. This process have now successfully been transferred to true roll-to-roll process using slot die coating, which is suited for high volume production and low cost (See Figure 15).

Figure 19. True roll-to-roll manufacturing equipment for electrodes and membranes.

The new roll-to-roll manufacturing equipment are also used for manufacturing of membranes.

The performance of MEAs with the new electrodes and membranes has been tested substantially with both hydrogen and wet reformate (See Figure 16).

The performance and variation of the new MEAs are truly State of the Art. Further efforts on durability is on-going.



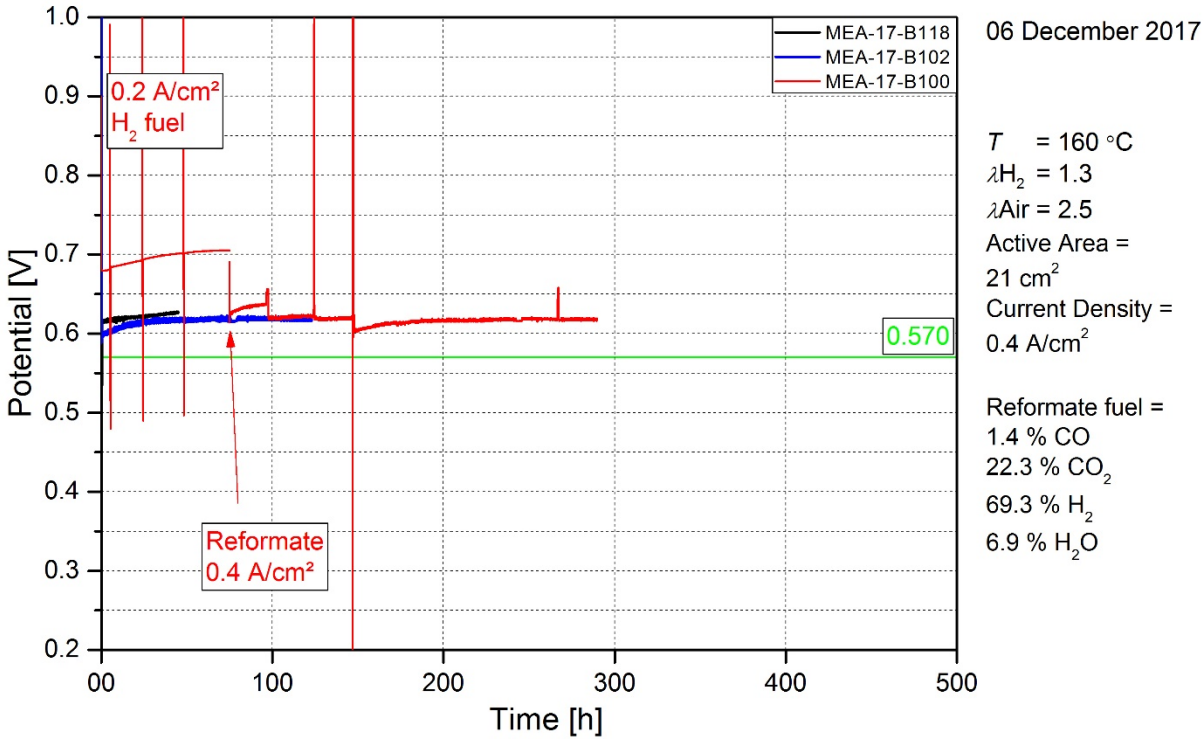


Figure 20. Performance and durability of the MEAs manufactured from materials produced on the roll-to-roll equipment.

1.4.4.1 MEA fabrication by hot-pressing

As part of the PhD project at DTU Energy the MEA effect of the hot-pressing process was investigated. Hot-pressing is a procedure to assemble the MEA. Electrodes and membrane are hot-pressed to ensure good ionic contact and to ease the subsequent handling of the MEA. Hot-pressing is done ex-situ before the cell is fixed in the test housing or stack. Alternatively, the MEA can be fixed in the test housing without prior hot-pressing. When the cell nuts are tightened and the cell is heated before test it makes it up for some degree of hot-pressing depending on the torque applied.

Numerous cells with and without hot-pressing were tested in a Baltic fuel cell fixture where the tension on the electrodes could be controlled. Cell performance at different compression is plotted in Figure 21 and Figure 22. It is evident that the cell compression has a strong effect on the performance when the MEA is assembled without hot-pressing (Figure 21). It seems to be the linear part of the voltage curve that changes slope. The slope gives an estimate of the series (ohmic) resistance in the cell, which is normally dominated by the membrane resistance and contact resistances. In this case it can be expected that it is the contact resistances that varies decisively. The best performance (highest voltage) is obtained with the higher cell compression force.

In the experiments where the cells were hot-pressed prior to fixation, the difference in performance as a function of cell compression in the cell test fixture is almost negligible and the performance is superior to the cells made without hot-pressing. The measurements are compared in Figure 23.

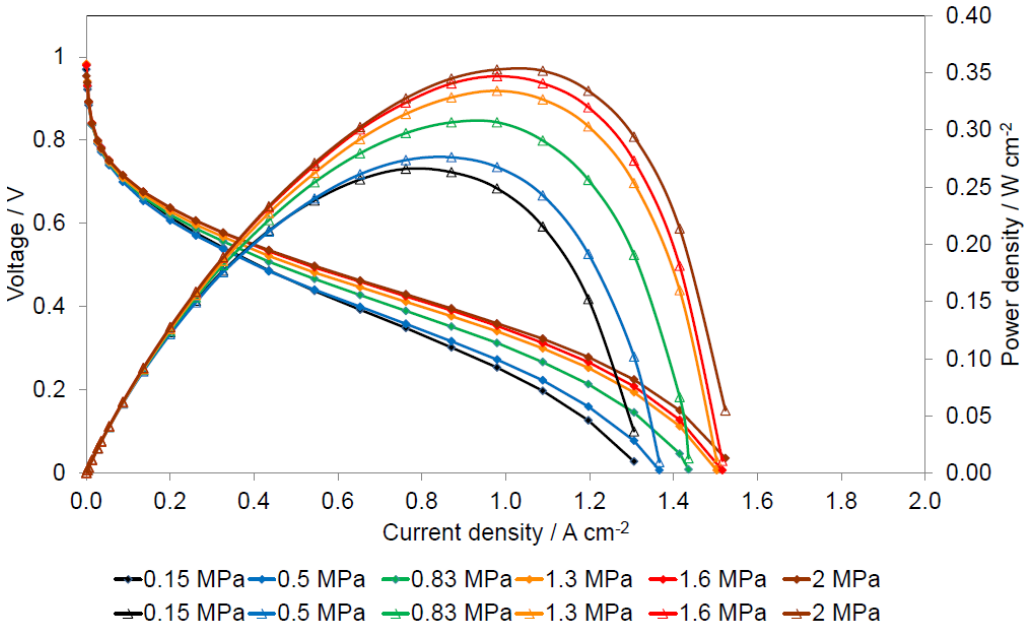


Figure 21. Effect of compression on MEA fabricated without hot press under air flow.

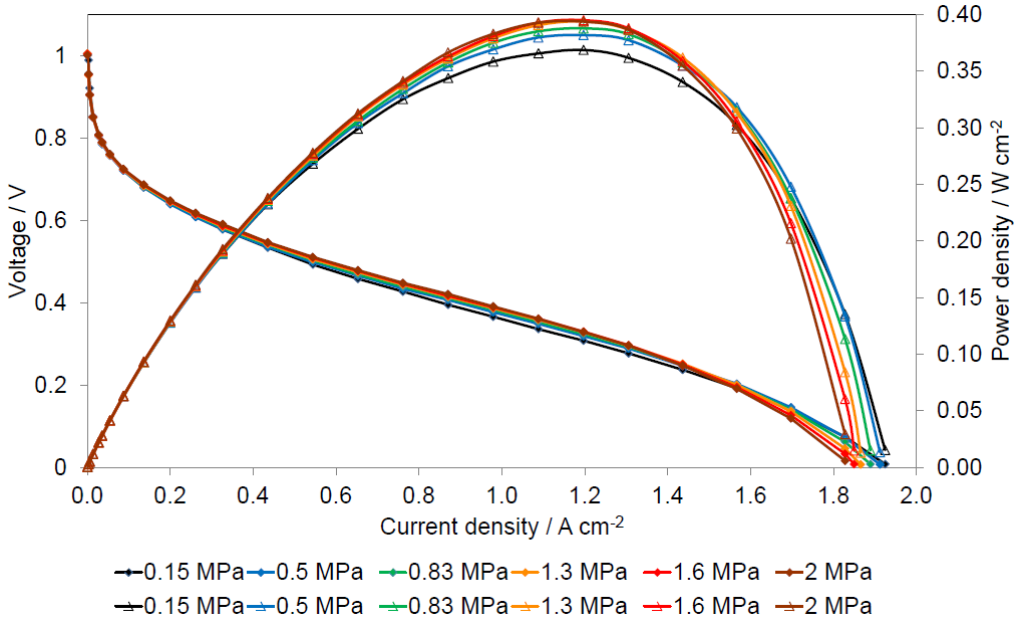


Figure 22. Effect of compression on MEA fabricated with hot press under air flow.

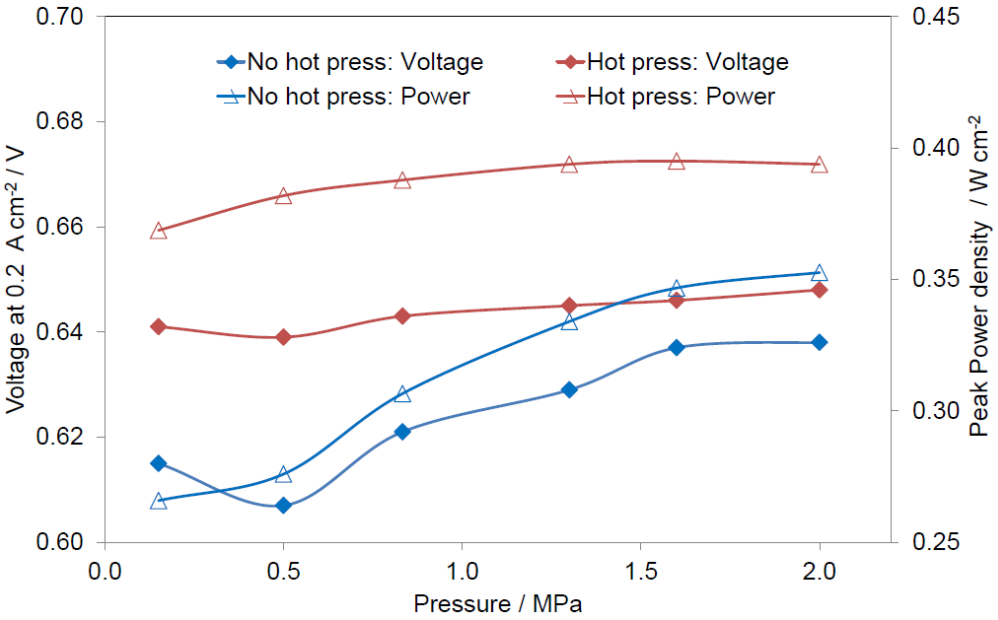


Figure 23. Voltage at 0.2 A cm-2 and peak power density as a function of contact pressure of cells operated under air flow.

Besides the polarization curves of the cell manufactured the two ways, Cross sections of the cells were examined for phosphoric acid distribution (Figure 24). The acid in the Hot-pressed MEA (*ex-situ*) is squeezed out into the GDL to a much higher extent than in the MEA assembled without hot-pressing (*in-situ* fabricated). This is not likely part of the explanation of the differences in series resistance, but it probably plays a role in the activation process where the cell gains performance over a quite long time in the beginning of life – often several hundred hours. The general understanding is that this is a process of acid redistribution.

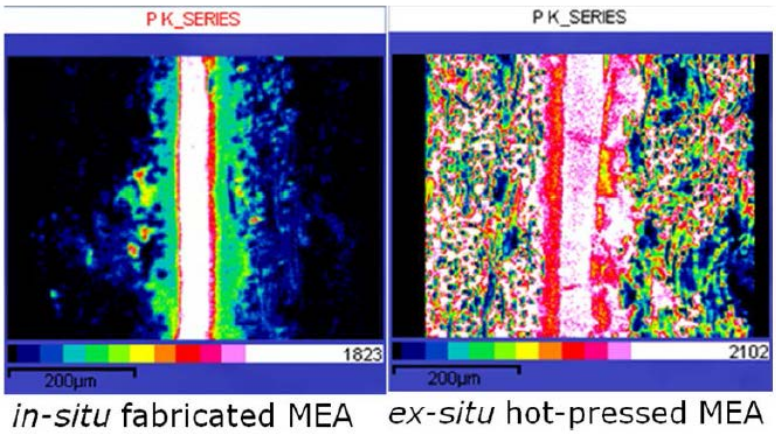


Figure 24. Phosphorus content analysis for cross sections of pristine samples of in-situ fabricated and ex-situ hot-pressed MEA and its corresponding acid distribution

1.4.4.2 Durable membrane

A new modified membrane was developed in the PhD project and tested for up to 10,000 hours at 180 °C. The high temperature, as compared to the usual 160 °C, was chosen to accelerate degradation. The voltage decay rate after activation was improved significantly with the membrane modification. Figure 25 shows the difference in decay rate in a 10,000 h durability test (one of the cells with a normal membrane was only operated for 5,000 hours).

The improvement was very significant and at 180 °C, decay rates as low as 2.7 µV h^{-1} is exceptionally low. Had the decay rate been for a cell operated at the usual 160 °C, this decay rate would still be considered very low.

DTU Energy has filed an internal patent notification, which is the first step in a patenting process. The novelty of the invention has been confirmed by an initial screening and DTU will file a patent application prior to the defence of the PhD study early 2018. For this reason, the invention cannot be disclosed and described in the final Smart MEA report. Refer to the PhD thesis of Arvind Kannan which will be held confidential until the PhD defence takes place. The modified membrane is considered one of the important results of the project.

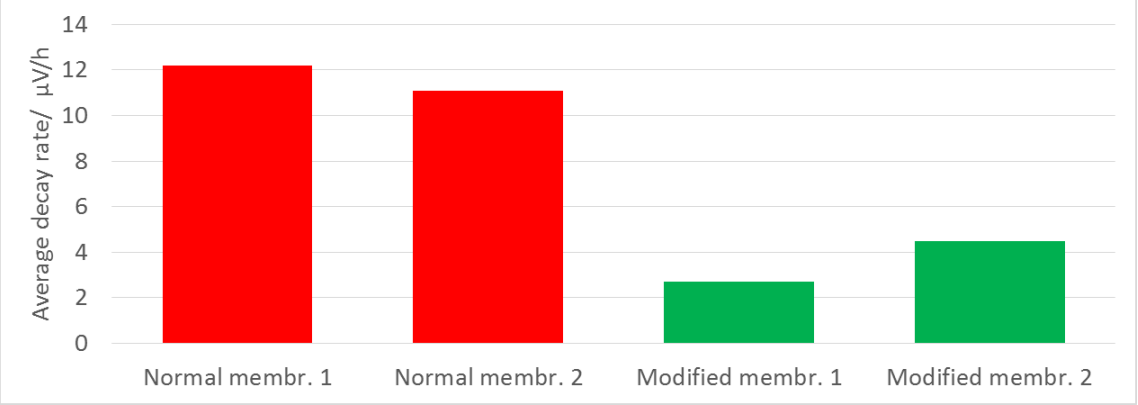


Figure 25. Decay rates over several thousands of hours at 180 °C for test cells with two normal PBI membranes and two modified membranes.

1.4.4.3 Stack test

In the project description, it was planned to make a short stack based on the stacking technology developed in previous ForskEL projects by partner IRD fuel cells (now EWII fuel cells). The purpose in this project was that DTU Elektro should perform stack testing to provide unbiased test results and to buid up stack test expertise. This task was not fulfilled because it was discovered that it required more preparations to furnish DTU Elektro with the necessary test infrastructure than originally anticipated.

1.4.5 WP 5. Dynamic tests and evaluation

(Lead: DPS)

The test facility at DPS has been updated (See Figure 18) in order to enable dynamic testing.

Figure 26. Test facilities at DPS.

The protocol for the dynamic test is shown in Table 3.

Table 3. Protocol for dynamic test. The test can be performed with both H_2 and reformat.

Start-up	1) Heat to 120^{o}C 2) Draw 100 mA cm^{-2} under λ_{H_2}=1.2 and λ_{Air}=2.0
Break-in	1) Heat to 160^{o}C 2) Draw 200 mA cm^{-2} under λ_{H_2}=1.5 and λ_{Air}=2.5 for 48h
Operation	1) Heat to 160^{o}C 2) Draw 250 mA cm^{-2} for 3h under λ_{H_2}=1.2 and λ_{Air}=2.0 3) Heat to 180^{o}C 4) Draw 550 mA cm^{-2} for 2h under λ_{H_2}=1.2 and λ_{Air}=2.0 5) Cool to 160^{o}C 6) Draw 250 mA cm^{-2} for 3h under λ_{H_2}=1.2 and λ_{Air}=2.0
Shut-down	1) Draw 100 mA cm^{-2} until T=120^{o}C 2) Stop flow (λ_{H_2}=0 and and λ_{Air}=0) and cool down to 80^{o}C
Conditions	1) Feed: H_2 / Air 2) Reactants are not humidified 3) Pressure is ambient

The results for standard MEAs are shown in Figure 19. The dynamic testing of MEAs based on PtCo/C is on-going.

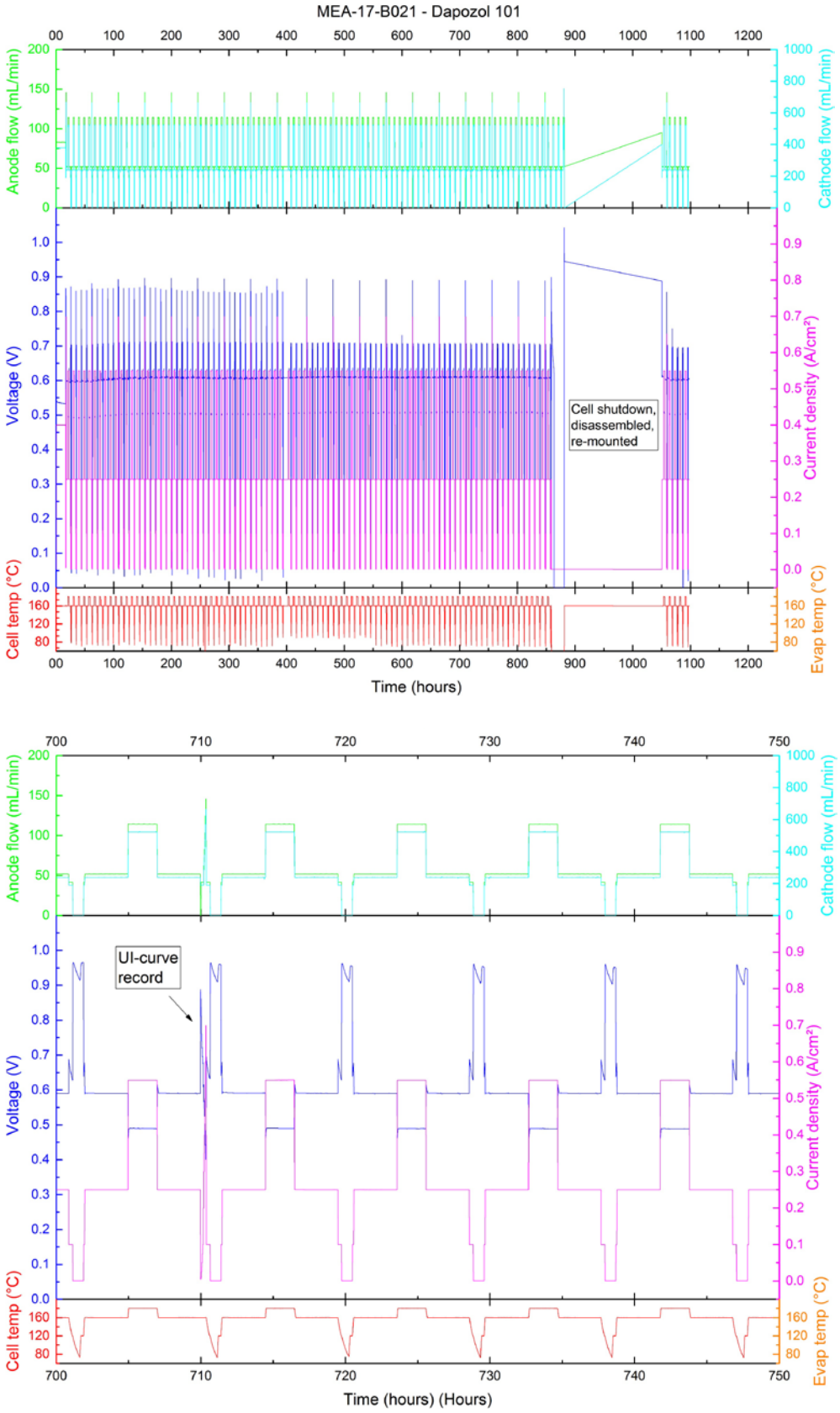


Figure 27. Results from dynamic testing of standard MEAs using Pt/C catalyst (1.5 mg/cm² Pt on both anode and cathode). The lower graph is a zoom-in on a few cycles.

Dynamic testing was performed at DTU Energy as well in collaboration with DPS. A detailed report was made in relation to a Master study [5]. The cells were subjected to the dynamic test sequence in Table 3 and the voltage profile at 250 and 550 mA cm^{-2} are plotted in Figure 28. The decay rates are listed in Table 4.

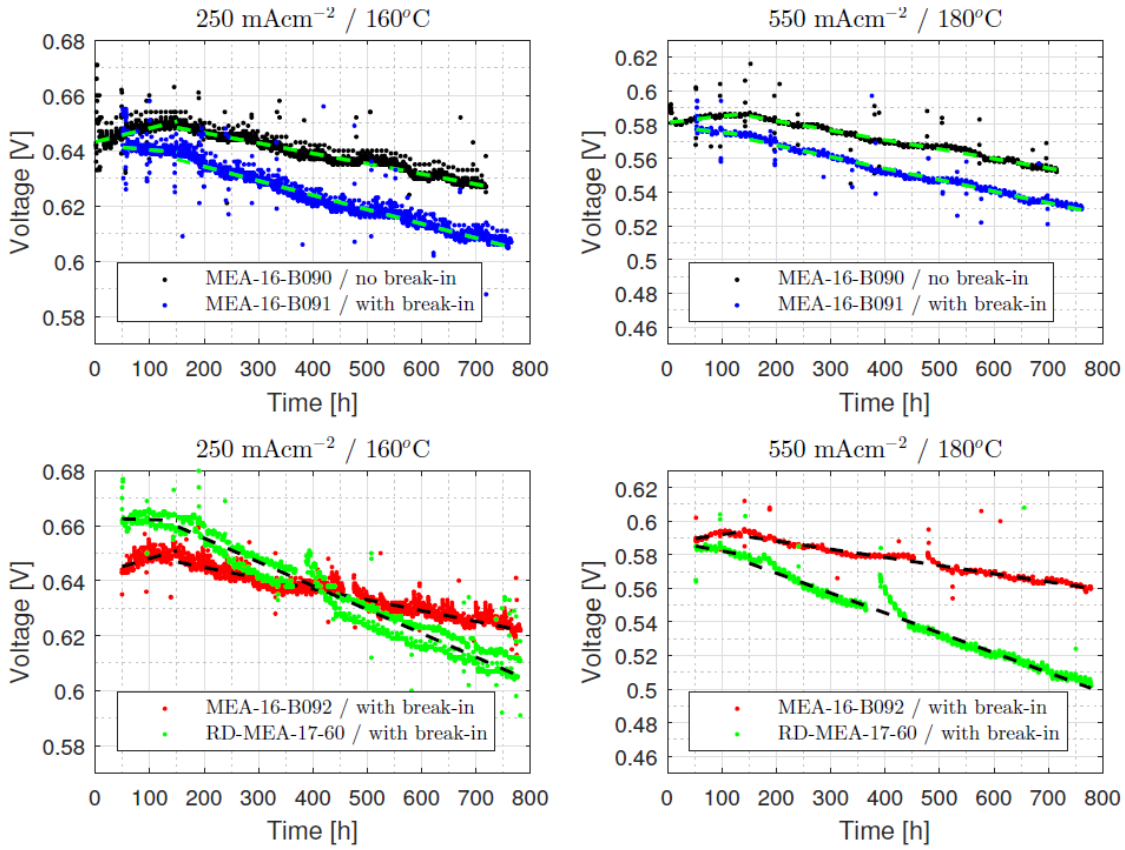


Figure 29. Selected examples of voltage development of cell from DPS tested at DSTU Energy with and without activation (break-in) [5]

Table 4. Degradation rates obtained by linear fitting of the voltage data and statistics of the fit's goodness (R^2).[5]

		Degradation Rate [μV/h]	Degradation Rate [μV/h]	Adjusted R^2 [-]	Adjusted R^2 [-]	Operation Point
		(< 150 h)	(> 150 h)	(< 150 h)	(> 150 h)	
MEA-16-B090	●	-49.54	37.22	0.9857	0.9925	
MEA-16-B091	●	17.69	51.80	0.9873	0.9906	**250 mA·cm^{-2}**
MEA-16-B092	●	-58.02	39.37	0.9652	0.9864	**160^{o}C**
RD-MEA-17-60	●	5.68	86.60	0.9763	0.9798	
		(< 150 h)	(> 150 h)	(< 150 h)	(> 150 h)	
MEA-16-B090	●	-37.07	55.95	0.9784	0.9945	
MEA-16-B091	●	36.01	68.27	0.9702	0.9944	**550 mA·cm^{-2}**
MEA-16-B092	●	-48.14	48.59	0.7379	0.9936	**180^{o}C**
RD-MEA-17-60	●	59.97	118.48	0.9704	0.99	

[5] Eftychia Bompolaki. Durability of High Temperature Proton Exchange Membrane Fuel Cells under Dynamic Operation with Start-up/Shutdown. Master project at DTU Energy, 2017.

1.4.6 Deliverables

Table 5. Overview of deliverables

Deliverable	**Status**	**Comment**
D0.1: Project internal website (M3, DTU-ECS)	Completed	
D0.2: Periodic reports (March 15th and Sept.15th, DTU-ECS)	Completed	The timing somewhat altered.
D0.3: Scheduling of a SmartMEA event (DTU-ECS M12)	Completed	Scheduled for November 2017.
D0.4: SmartMEA event (DTU-ECS, timing TBD)	Completed	Held in collaboration with a other fuel cell projects in the framework of Danish Electrochemical Society November 2017
D1.1: Specifications of criteria and targets of SMART MEAs (M20, DPS)	Not accomplished	A detailed study was performed at DTU Elektro, but time was unfortunately not sufficient at the end to establish smart grid related protocols.
D1.2: Input for MEA test protocols (M24, DTU-ECS)	Completed	Protocol used for dynamic testing
D1.3: System analysis and simulation report (M36, DTU-Electro)	Completed	Report “Robust HT-MEAs for dynamic operation under smart grid conditions”, 2017
D2.2: Systematic characterization results of membranes (M26, DTU-ECS)	Completed	Cannot be discussed here due to patenting

1.4.7 Milestones

Table 6. Overview of milestones

Milestone	**Status**	**Comment**
M0.1: Recruitment of PhD student (M3, DTU-ECS)	Completed	PhD student submitted thesis, November 2017
M0.2: Annual project review (DTU-ECS M12, M24)	Not as a formal procedure	Not carried out as a formal procedure, but focus and priorities were adjusted when needed during the project.
M0.3: Final report (M36, DTU-ECS)	Completed with this report	This report.

M1.1: Requirements for dynamic operation of SMART MEAs (M16, DTU-Elektro)	Completed	In report "Robust HT-MEAs for dynamic operation under smart grid conditions", 2017
M2.1: Strategy for electrode improvement (M6, DTU)	Completed	Focus on the role of the GDL and the acid retention
M2.2: First electrodes samples for MEA test (M20, DPS)	Completed	New electrodes were tested at both DTU Energy and DPS
M2.3: Final evaluation of next generation electrodes (M30, DTU)	Completed	Electrodes developed were also tested.
M3.1: Strategy for enforcement (M3, DTU ECS)	Not Completed	Enforcement was not addressed to the extent originally planned for due to other priorities.
M3.2: New MEA design (M26, DTU ECS)	Completed	New design by new way of making the membrane. Cannot be discussed here due to patenting.
M4.1: First short stack with IRD components (M9, DTU ECS)	Skipped	I made little sense to build up the extensive text infrastructure at DTU Elektro.
M4.2: Optimized MEA assembly process that can be utilized with the membrane and electrodes developed in WP3 and WP4(M30, DPS)	Partly completed	Assembly process studied and lessons learned. However, the MEA assembly process has not changes
M5.1: Test Protocol for Smart grid MEAs (M30, DTU-ECS)	Not accomplished	A detailed study was performed at DTU Elektro, but time was unfortunately not sufficient at the end to establish smart grid related protocols.

1.4.8 Dissemination of the project

Result of the project were disseminated at multiple occasions

1.4.8.1 Conventional scientific dissemination

8 papers were published or submitted for publication in **international peer reviewed journals.** Two or more additional papers are in the pipeline (on MEA assembly and on the membrane to be patented).

1 **patent application** on an improved membrane technology is being processed at DTU. The application will be filed during the initial months of 2018.

A total of 25 **conference presentations** were so far made with credit to the project. Several additional presentation are planned or expected. On 26 - 27 Septem-

ber 2018, DTU Elektro - CEE organizes the 7th International Conference on Renewable Power Generation, RPG 2018 [6] where the aims is to have a special session on “Accelerating the onset of hydrogen economy by using hydrogen solutions for grid services”. DTU Energy will participate in World Hydrogen Energy Conference (WHEC) 2018 in Rio de Janeiro and present work on cell durability and acid retention.

The project will moreover be summarized in the Danish magazine BioPress after the final report.

1.4.8.2 Theses and reports

2 **PhD theses** were made. One PhD project (A, Kannan) was fully financed by the project. The thesis was submitted November 2017 and was waiting for the evaluation at the time of reporting. The other PhD project (A. Pensini) was only partly financed by the project. After submission the thesis was not approved for defence and rewriting was required.

1 **Master thesis** on dynamic testing was completed. The student was later hired for reenrolment as a PhD student to further pursue the path for electrospinning

1 Comprehensive **technical report** on the fuel cell study at DTU Elektro

1.4.8.3 Designated project events

Electrochemical Science and Technology Conference 2017 and Annual Meeting of The Danish Electrochemical Society, 2-3 November

(Special theme this year: Low-temperature fuel-cell and electrolysis technologies)
A two day fuel cell and electrolyzer conference was arranged with the Danish Electrochemical Society [7] as host and with a number of national projects as co-organizers (including SmartMEA). The aim was to provide an overview of the many activities in Denmark in the fields of fuel cells and electrolyzers. Four invited speakers from Germany, South Korea, Switzerland and Denmark took part. SmartMEA was presented with a poster.

Den Danske Brint- og Brændselscelledag 2017 (Danish Hydrogen and Fuel Cell Day)
This event was arranged 28 November at University of Southern Denmark by Hydrogen Denmark in collaboration with Innovation Fund Denmark, EUDP and University of Southern Denmark. SmartMEA was presented with a poster.

[6] RPG 2018: http://events.theiet.org/rpg/index.cfm

[7] http://www.electrochemistry.dk

1.5 Utilization of project results

1.5.1 Utilization in general

The project has advanced the HT-PEMFC technology significantly. DPS has expanded significantly over the latest years. The staff has more than doubled and the space for production has been extended. The technical development and knowledge gained will be of great importance to the company. The nature of the accomplishments of the project is technical and thus highly relevant to the general gearing up of DPS. The findings will certainly be utilized in the commercialization of HT-PEMFC for which DPS is a leading player.

At DTU Energy the results and new ideas of the project has triggered new pathways for research and development that will be pursued. The work on phosphoric acid movement and retention resulted in quite unexpected findings and a new Master project will be initiated together with DPS on the understanding of the activation process in the cells. We believe that activation is closely coupled to acid redistribution, but also to hot-pressing conditions. It is also possible that the new membrane technology developed during the project can shorten the time for activation.

Electrospinning is a hot topic internationally and in the project we have started several activities without being able to take it all the way to cell tests yet. This will a major activity at DTU Energy in the near future. The electrospinning specialist Wen-Jing Zhang, who contributed to the project has now joined the section Proton Conductors full time. Two new PhD projects and a postdoc project on electrospinning will start early 2018 at DTU Energy. The candidates are found.

On a DTU level this project has helped CEE/DTU Elektro into a closer and better collaboration with DTU Energy, e.g. leading to a successful proposal in the H2020 and EUDP programs.

Results from the project has led to increased CEE/DTU Elektro activity in the field of integrated energy systems. As consequence CEE has successfully engaged in new activities in the field between electric power, heat and gas. CEE has currently several PhD students working with integrated energy infrastructure.

The center participates in a H2020 project QualyGridS [8] aimed at providing grid services using water electrolyzers and a EUDP project SPGC [9] aimed at providing back-up power for grid substations by use of fuel cells. The expertise developed in conjunction with and in other parallel activities has enabled CEE to participate in the QualyGridS project as a main driver of a technical report on the potential for water electrolyzers to providing grid services within Europe [10]. Similarly, the experience developed during this and other subsequent fuel cell/electrolyzer projects has resulted in an increased interest from Chinese stakeholders to approach CEE which as lead to a bi-lateral collaboration with Chinese partners.

[8] QualiGridS H2020 project: www.qualygrids.eu

[9] SPGC EUDP project – just kick started December 2017.

[10] S. You et al. Technical report, QualyGridS project: “Electrical Grid Service Catalogue for Water Electrolyser (D1.1), Nov. 2017.

1.5.2 Patents

The developed membrane technology that is only mentioned and not described in this report will be patented by DTU. The process has been long because the verification of the increased durability has taken up to 10,000 hours. The section Proton Conductors at DTU Energy has moved the entire lab to a new building during November 2017 and all durability experiment have been stopped. It is the plan do start e new series of cells with the new membrane early 2018 when the durability setups are operational again. Shortly before the PhD defence when the invention will be exposed, the patent application will be filed.

1.5.3 On the business plan of Danish Power Systems

The various cost scenarios for HTPEM FCs in relation to CHP applications have been analysed both in EU and in US. Table 7 shows the EU target and Figure 30 shows the US targets.

Table 7. Comparison of mid-size fuel cell installation KPI's given in the FCH-JU MAWP for complete CHP systems. [11]

KPI	UNIT	SOA	FCH-JU TARGETS		
		2012	2017	2020	2023
CAPEX	€/kW	6,000 - 10,000	5,000 - 8,500	4,500 - 7,500	3,500 - 6,500

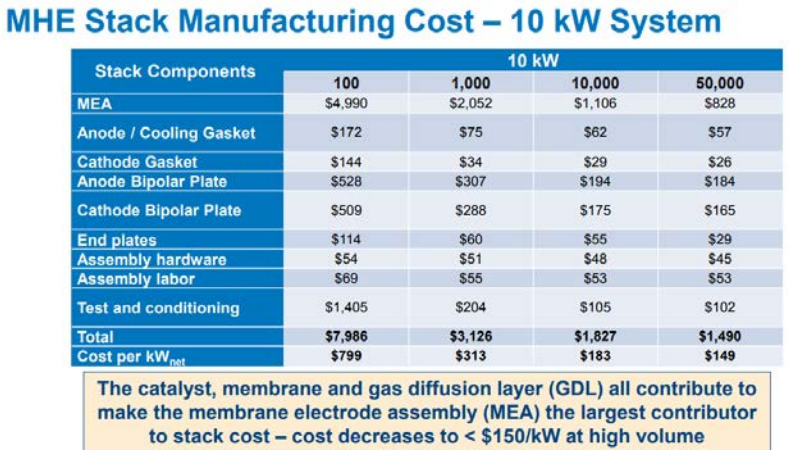


Figure 30. Cost targets for stacks in relation to CHP systems estimated by Dept. of Energy (US). [12]

[11] http://www.fch.europa.eu/sites/default/files/documents/FCH2%20JU%20-%20Multi%20Annual%20Work%20Plan%20-%20MAWP_en_0.pdf

[12] https://energy.gov/sites/prod/files/2017/01/f34/fcto_webinarslides_mfg_cost_analyses_fc_nontransport_apps_012417.pdf

The cost of the MEA is a significant part of the entire CHP system (~50%), meaning that it is crucial to achieve a low MEA cost in order to obtain a commercially viable CHP solution based on HTPEM FCs.

The activities with SmartMEA and related projects have addressed these challenges in terms of both cost analysis and manufacturing processes.

A detailed cost analysis for large scale MEA production have been performed by DPS. The result is that the MEA cost requirements for 1000 stacks (10 kW) can be met.

In conclusion, the results obtained in the SmartMEA project have made a significant contribution to the commercialization of HTPEM for Smart Grid applications like CHP.

1.5.4 Teaching

Results from the PhD work was used in the DTU course 31778 “Emerging Energy Technologies” where fuel cell based combined heat and power technology as well as storage technologies were introduced in the portfolio of distributed energy technologies. DTU Elektro - CEE has several PhD students working with integrated energy infrastructure.

The Course 47302 “Hydrogen and Fuel Cell Chemistry”, is a laboratory course at DTU Energy, involving multidisciplinary work with fuel cells and electrolyzers. The PhD students take part in the teaching there. It may moreover be possible to include parts from SmartMEA after the next run in January.

1.6 Project conclusion and perspective

1.6.1 Conclusion

The project is considered successful by the participants. The HT-PEMFC technology has reached a rather mature state of development – among other things thanks to several projects with ForskEL. These project have been addressing different issues at different states of the development. SmartMEA has been the most focused so far on the issue of robustness and manufacturability (the DuraPEM projects should be mentioned here too and SmartMEA has benefitted from synergy with the latest, DuraPEM III). This is natural as DPS has developed the manufacturing capability tremendously during the same period. The activities and results of SmartMEA has contributed to directing that towards the best possible.

The main technical results to list in the conclusion are:

1. New insight in the unexpected effect of the choice of gas diffusion layer for the electrodes.
2. A new membrane technology which is expected patentable
3. Platinum leading has been reduced
4. Electrospinning has been initiated for GDL manufacture
5. Dynamic cell testing has been started

6. A test protocol for dynamic testing has been made
7. Effects of cell activation as compared to non-activation have been revealed
8. An extended study of the application of the HT-PEMFC has been carried out and reported

1.6.2 Perspectives

Some of the detailed perspectives were presented in the previous chapter on utilization. Here the broader perspective will be addressed.

HT-PEMFC has been a steady activity in Denmark since the idea was conceived at Case Western Reserve University in Cleveland, Ohio. Today two of the leading HT-PEMFC industries are located in Denmark (Danish Power Systems and Serenergy) and the area has been granted many R&D projects. Both industries are now experiencing steady growth and may be some of the first fuel cell companies that actually make a revenue. One important key to a commercial success is the fact that the HT-PEMFC technology does not rely on a hydrogen infrastructure, but can be operated on conventional fuels via a reformer. Traditional low temperature PEMFC (LT-PEMFC) technology can in principle do the same, but the system is then much more complicated. The number of commercial players on HT-PEMFC (as well as research groups) is much more limited than for LT-PEMFC and Denmark has a very strong position.

The support from the different research and development programmes have played a decisive role in this development and it is possible that the HT-PEMFC will experience real commercialization since it targets applications that do not require additional infrastructure and do not depend on subsidies (telecom, range extenders etc. all on methanol).

The primary challenges for fuel cells today are cost and durability and there is still much work to do to improve the technology on both issues. The project SmartMEA has been a significant stepping stone in that development and beside the technical accomplishments, it has set several new directions for improvements and better understanding.

The role of HT-PEMFC in a smart grid scenario is perhaps not completely mapped by this project, but hopefully, is has contributed to that as well.

1.7 Annex 1. Publications

The listed publications are made under SmartMEA or with contributions from SmartMEA.

International peer reviewed journals

9. T. Søndergaard, L. N. Cleemann, H. Becker, D. Aili, T. Steenberg, H. A. Hjuler, L. Seerup, Q. Li and J. O. Jensen. Long-term Durability of HT-PEM Fuel Cells Based on Thermally Cross-linked Polybenzimidazole. J. Power Sources **342**, 570-578 (2017)
10. H. Becker, L. N. Cleemann, D. Aili, J. O. Jensen, Q. Li. Probing phosphoric acid redistribution and anion migration in polybenzimidazole membranes. Electrochem. Commun. **82** 21–24 (2017)
11. M. T. D. Jakobsen, L. N. Cleemann, L. Zhong, H. Becker, T. Steenberg, H. A. Hjuler, L. Seerup, Q. Li and J. O. Jensen. Catalyst Degradation under Accelerated Stress Test for PBI based HT-PEM Fuel Cells – Effect of Humidification. Published online in Electrocatalysis (2017).
12. S. B. Simonsen, Y. Wang, J. O. Jensen and W. Zhang. Coarsening of carbon black supported Pt nanoparticles in hydrogen. Nanotechnology **28** 475710- (2017).
13. A. Kannan, Q. Li, L. N. Cleemann, J. O. Jensen. Acid distribution and durability of HTPEM fuel cells with different electrode supports. Accepted for publication in Fuel Cells
14. M. T. D. Jakobsen, L. N. Cleemann, H. Becker, T. Steenberg, H. A. Hjuler, L. Seerup, Q. Li, J. O. Jensen. Parametric Accelerated Stress Test Study in Relation to Long-term Durability of PBI-based HT-PEM Fuel Cells. Submitted to J. Electrochem soc.
15. A. Pensini, C.N. Rasmussen, W. Kempton, Economic analysis of using excess renewable electricity to displace heating fuels, Appl. Energy. **131** (2014) 530–543. doi:10.1016/j.apenergy.2014.04.111.
16. A. Pensini, M. Robinson, N. Heine, M. Stadler, A. Mammoli, Grid-friendly scheduling of distributed energy resource clusters via real-time pricing. Submitted to IEEE - Transaction on Sustainable Energy, 2015.

Patent applications

1. A Kannan, J. O. Jensen, D Aili, Q. Li and L. N. Cleemann. Title confidential. Invention: more durable membrane. Internal patent notification at DTU. Novelty approved. The patent office at DTU will file a patent application.

Conference presentations

1. J. O. Jensen, L. N. Cleemann, Q. Li, M. T. D. Jakobsen, H. A. Hjuler and T. Steenberg. Status on durability of high temperature PEMFC. 20th World Hydrogen Energy Conference (WHEC2014). June 15-20. 2014, Gwangju, South Korea (oral by Cleemann).
2. A. Vassiliev, S. Martin Fernandez, K. B. Andersen, L. N. Cleemann and J. O. Jensen. Methods for HT-PEM FC electrodes preparation and their influence on MEA activity. 4th Carisma Conference. December 1-3. 2014, Cape Town, South Africa (poster)

3. Q. Li. High Temperature PBI based PEMFC - Status, Challenge and Recent Progress. Presentation at EEST, 2014-10-31, Shanghai, China
4. J. O. Jensen. L07 - Catalyst layers. 4M HT-PEM Summer School. Schæffergården, Gentofte, 24 – 28 of August 2015.
5. J. O. Jensen, S. Martin, A. Vassiliev, L. N. Cleemann and Q. Li. The effect of hydrogen pressure on the tolerance for co of high temperature PEM fuel cells. Annual Meeting of Danish Electrochemical Society. University of Copenhagen, 1-2 October, 2015 (abstract, oral)
6. H. A. Hjuler, R. Kerr, T. Steenberg, C. Terkelsen, J. O. Jensen, L. N. Cleemann and Q. Li. Advanced Materials for High-Temperature PEM Fuel Cells. The 3rd International Conference on Advanced Electromaterials (ICAE2015). Jeju, South Korea. November 17-20, 2015
7. J. O. Jensen, Y. Hu, L. Zhong, L. N. Cleemann and Q. Li. Challenges in Membrane Electrode Assembly Technologies. 2nd APT-STEP Workshop on Novel Material Technologies for Alternative Powertrains. Thessaloniki, Greece, 25-26 February 2016 (invited plenary talk),
8. J. O. Jensen, M. T. D. Jakobsen, L. N. Cleemann, T. Steenberg, H. A. Hjuler and Q. Li. Systematic Study of Durability of High Temperature PEM Fuel Cells at Selected Temperatures, Flow Rates and Loads. Electrochemical Society fall meeting, Prime 2016, Honolulu, US, October 2-7. 2016 (oral)
9. A. Vassiliev, L. N. Cleemann, Q. Li, and J. O. Jensen. Hydrogen Sulfide Tolerance in High Temperature PEMFCs. Electrochemical Society fall meeting, Prime 2016, Honolulu, US, October 2-7. 2016 (oral by Vassiliev)
10. T. Steenberg, H. A. Hjuler (Danish Power Systems), J. O. Jensen, and Q. Li. Optimization of Catalyst Layer Properties for High Temperature Polymer Fuel Cells. Electrochemical Society fall meeting, Prime 2016, Honolulu, US, October 2-7. 2016 (oral by Steneberg)
11. J. O. Jensen, T. Søndergaard, D. Aili, L. N. Cleemann, T. Steenberg, H. A. Hjuler, H. Becker, L.Zhong and Q. Li. A Durability Study of High-temperature PEMFC. Effects of Operating Parameters and Thermal Curing. Fifth International Carisma Conference, Newcastle upon Tyne, UK, April 10-12, 2017 (invited keynote)
12. J. O. Jensen, T. Søndergaard, L. N. Cleemann, T. Steenberg, H. A. Hjuler and Q. Li. Durability Studies of High Temperature PEM Fuel Cells. Operational Parameters, Accelerated Testing and Acid Retention. 6th European PEFC and Electrolyzer Forum, Lucerne, Switzerland, 4-7 July 2017 (oral).
13. S.B. Simonsen, Y. Wang, J.O. Jensen, W. Zhang. In situ TEM study of the coarsening of carbon black supported Pt nanoparticles in hydrogen. Electrochemical Society fall meeting, National Harbor, Maryland, US, October 1-5. 2017 (oral)
14. J. O. Jensen. Recent Progress in Durability of High Temperature PEMFC at DTU Energy. Workshop im Rahmen des Projektes QUALIFAX “Qualität von HT-PEM Brennstoffzellen”. Oldenburg, Germany, 5-6 December 2017. (invited talk)
15. A. Kannan, Q. Li, L.N. Cleemann, J.O. Jensen. “Long term Durability of High Tem-perature Polymer Electrolyte Membrane Fuel Cells based on Acid Doped Polybenzim-idazole” Poster presented at the 21st European PEFC and electrolyser Forum Con-ference, Lucerne, Switzerland, (2017)
16. A. Kannan, Q. Li, L.N. Cleemann, J.O. Jensen. “Durability of HTPEM fuel cells with different electrode supports” Poster presented at the 4th international CARISMA Conference, Newcastle, United Kingdom, (2017)
17. A. Kannan, S.Martin, Q. Li, L.N. Cleemann, J.O. Jensen. “Parametric design and De-velopment of HT PEM Fuel cells” Poster presented at the Danish Electrochemical So-ciety conference at The Technical University of Denmark, Lyngby, Denmark (2017)

18. A. Kannan, L.Seerup, Q. Li, L.N. Cleemann, J.O. Jensen. “Development of HTPEM fuel cells for smart grids” Poster presented at The Danish Hydrogen and Fuel cell day at the University of South Denmark, Odense, Denmark (2016)
19. A. Kannan, L.Seerup, L.N. Cleemann, Q. Li, J.O. Jensen. “Acid management of HT PEM Fuel cells” Poster presented at the PhD symposium arranged by DTU Energy at the Technical University of Denmark, Lyngby, Denmark, (2016)
20. A. Kannan, S.Martin, Q. Li, L.N. Cleemann, J.O. Jensen. “Development of HT PEM Fuel cells” Poster presented at the PhD symposium arranged by DTU Energy at the Technical University of Denmark, Lyngby, Denmark, (2015)
21. Q. Li. “Development of HTPEM fuel cells for smart grids” Poster presented at The Danish Hydrogen and Fuel cell day at the University of South Denmark, Odense, Denmark (2017)
22. A. Pensini, E. Larsen, Bidding strategy for a wind power producer with fuel cell integration, submitted to IEEE - PES Innovative Smart Grid Technol-ogies Latin America Conference 2015, extended abstract – accepted.
23. A. Mammoli, A. Pensini, Can greedy customers be good citizens? Coor-dinated distribute energy resource optimization via power flow simulation, 6th Int. Conf. on Integration of Renewable and Distributed Energy Resources (IRED), poster (2014).
24. A. Pensini, M. Robinson, N. Heine, M. Stadler, A. Mammoli, Assessment of grid-friendly collective optimized scheduling, submitted to IEEE - PES Inno-vative Smart Grid Technologies Asian Conference 2015.
25. A. Pensini, A. Mammoli, Feasibility of optimizing large utility electricity customers on a distribution feeder, published in Proceedings of CYSENI 2014. 2014. p. 112-119., (2014).
26. J. O. Jensen, T. Søndergaard, A. Kannan, D. Aili, L. N. Cleemann, H. Becker, T. Steenberg, H. A. Hjuler, Q. Li. Durability of High-Temperature PEM Fuel Cells. 22nd World Hydrogen Energy Conference (WHEC2018). Rio de Janeiro, 17-22 June, 2018 (abstract)

PhD theses

1. PhD thesis. Arvind Kannan, Parametric development and durability of HTPEM Fuel Cells. DTU, November 2017. Submitted.
2. Alessandro Pensini, PhD report on “Energy Systems Integration and Storage”, 2015 (*not yet approved for the PhD defence*)

Other publications

1. Eftychia Bompolaki. Durability of High Temperature Proton Exchange Membrane Fuel Cells under Dynamic Operation with Start-up/Shutdown. Master project at DTU Energy, 2017.
2. A. Pensini et al, report on “Robust HT-MEAs for dynamic operation under smart grid conditions”, 2017. (The final report of the SmarMEA study at DTU Elektro)