FINAL REPORT for

EUDP08-1:
"Development of Low Cost and High performance MEA - and bipolar plate processes"

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Summary

The objectives for the total 36-month project have been to develop:

- MEA Laminate Fabrication Processes: Develop 7-layer MEA laminate materials and fabrication routes suitable for low-cost series production, through state-of-the-art knowledge and by using powder coating processes and roll-to-roll laminate processes.

- Bipolar Plate Fabrication Processes: Develop materials and fabrication routes suitable for low-cost series production of the bipolar plates, by careful material selection through state-of-the-art knowledge, e.g. focus on carbon composite bipolar plates made using thermoplastics or thermoset resins.

The project was divided into two phases. The report covers both phases. The milestones accomplished through the project lists also the results of the project:

- Specifications of MEAs and MEAs processes

- Construction of a MEA manufacturing line based on dry electrode powder

- Specifications of bipolar plates and process for making bipolar plates

- Construction of a graphite bipolar plate manufacturing line

- Development of seal material for LT and HT-PEM application

- Testing of MEAs made on the line verifying the line and the MEAs

- Testing of graphite bipolar plate made on the line verifying the line and the plates

- Price reduction of flowplate cost

- Manufacturing and testing of DMFC stacks for demonstration

- Techniques for mould injection of seals developed

- MEA laminate for the stack test completed

- Materials composition for hot-pressing developed

- Materials composition for injection molding developed

- Production route for hot pressing defined

- Production route for injection-molded bipolar plates defined

- Bipolar plate production for the stack test completed

- Report on single-cell MEA test completed

- Report on bipolar plate test completed

- Test and evaluation of LT PEM & DMFC completed

- Evaluation of the MEA laminate and bipolar plate production cost at a future series production

The project has been completed with these important results for the project partners.

The Catalyst Coated Membrane production line has been established for production of PEM and DMFC MEA laminate.

The hot pressing flow plate production line has been established for production of molded graphite bipolar flow plates.

The MEA laminate process and the bipolar plate process together with the sealing materials have been finalized and verified in the last part of the project

The MEAs manufactured on the line meet the performance requirements relatively good.

The bipolar plates manufactured on the line meet the performance and cost requirements.

The PEM and DMFC stack prices are reasonably in accordance with the target values. An analysis of the stack price structure shows that the MEA catalyst cost together with production costs are the dominating price factor. Research and development effort in order to reduce the prices within the PEM fuel cell technology should be focused on reducing the amount of catalyst used in PEM Fuel Cell. and reduce production costs

The project has in overall terms achieved the technical and economical milestones.

Introduction

In terms of global energy and environmental problems fuel cells are expected to play a major role in the future energy-supply sectors. However the present manufacturing costs far exceed competitive prices and will fall significantly only when the supply of components and the manufacturing processes have been industrialized.

The project goals comply with targets defined in the national Danish strategy on PEM FCs [1)]. The two following overall objectives goals included in the Danish PEM FC strategy:

1. Support set-up of a Danish PEM FC technology comprising production of cells, stacks, and systems

2. Secure that the technology becomes a reel industrial activity where targets and finances are consistent and sufficiently attractive, for both scientists and industry, to continue PEM FC research and development work in Denmark.

[1)] Dansk PEM brændselscellestrategi (www.Partnerskabet/Strategier/Dansk PEM brændselscellestrategi)

The national target costs were defined at the beginning of the project as follows [2)]:

Year	LT PEM Stack (€/kW)	DMFC Stack (€/kW)	HT PEM Stack (€/kW)
2008	20,000	15,000	15,000
2012	3.100	9,000	3,000
2020	1.200	1,500	400

[2)] targets defined in the national Danish road maps on PEM FCs, latest revision April 2011

Innovative development with respect to state-of-the-art

FC stacks and their respective components are in the early stages of manufacturing. Manufactured using fabrication methods that have been scaled up in size, but do not incorporate high volume manufacturing methods. The MEA as an example was accomplished in steps as discrete operations with most of actual labor done by hand. To reduce the stack cost, high-volume and low cost electrode and bipolar plate fabrication processes were essential.

The aim of the present project was to combine the results obtained in earlier national and international projects and through component and manufacturing technology development obtain significant FC cost reduction and high performance. The fabrication processes developed in the present project have been targeting at meeting future requirements for mass-production capability.

The project have been taking benefit from a number of national and international projects, especially from two FP5 projects (PortaPower [ENK5-CT-2002-00669] & 50PEM-HEAP [NNE5-2001-882]) The results from these projects have been used and further developed in the present project. A by the piece dry MEA processing route, well suited for upscale to a continuous MEA-production was established in the PortaPower project and a mould-in-shape-bipolar plate fabrication route with a large cost reduction potential was established in the 50PEM-HEAP project. However, the partner responsible for the bipolar plate process has with drawn from this business area stopped the development and sold the technology to a third party, a big setback for the positive cost trend development. However the development through this project has overcome this challenge and succeed in establishing the bipolar plate production continuing the positive cost trend.

The objectives for the total project have been to develop:

1. **MEA Laminate Fabrication Processes:** Development of 7-layer MEA laminate materials and fabrication routes suitable for low-cost series production, through state-of-the-art knowledge and by using powder coating processes and roll-to-roll laminate processes.

2. **Bipolar Plate Fabrication Processes:** Development of materials and fabrication routes suitable for low-cost series production of the bipolar plates, by careful material selection through state-of-the-art knowledge, e.g. focus on carbon composite bipolar plates made using thermoplastics or thermoset resins.

By implementation of such a coating process it was expected that new routes for catalyst content reduction and high performance MEA laminates would be obtained. This would enable the partners to obtain the target goals defined for performance, catalyst content and cost in the national Danish strategy on PEM FCs. State-of-the-art 5-layer MEAs are comprised of a membrane and anode and cathode catalyst electrodes bonded to two gas diffusion media. The 7-layer MEA incorporates an elastomeric gasket edge seal system integrated into the 5-layer MEA.
The spray rolling technique is a simple, flexible, cheap and environmentally friendly process with the capability for mass production. An advantage of this fabrication route is that there are very few rheological requirements to the electrode mixture making it possible to refine the compositions, layer thickness and interface reducing the catalyst loading.

The target was to specify fabrication processes making it possible to produce high performance MEA laminates reducing the cost with 50% (achieved by reducing labor cost and catalyst amount) and bipolar plates at a price of 50 €/m^2 also for a low number of plates (5.000 plates).

Scientific and technological objectives

The main objective was to develop materials and fabrication processes for cost-competitive MEA (Membrane Electrode Assembly) laminates and bipolar plates for both HT-(High temperature and LT (Low Temperature) PEMFC (Proton Exchange Membrane Fuel Cells.

The main objectives have been reached by integrating results obtained in earlier national and international projects and through the R&D work described in the WPs listed below for the project :

WP0: Coordination and management. The project is coordinated by IRD from this WP. The work includes economic management of the project; communication with EFP; organize technical project meetings and compilation of progress reports.

WP1: Material & Process specification. The objective is to perform a material and process specification followed by a cost analysis of materials and fabrication techniques to be pursued within the project.

WP2: MEA laminate development. The objective is to develop optimized MEA laminates with reduced catalyst loading achieved by developing a cost effective roll-to-roll laminate process.

WP3: Bipolar plate development. The objective is to develop cheap and reliable material and fabrication routes for high quality graphite bipolar plates based upon carbon-thermoset and carbon-thermoplastic polymer composites by extrusion, pressing or injection molding techniques.

WP4: Single cell & prototype testing. The R&D work includes in single-cells test of both the MEA laminate compositions and bipolar plates developed and fabricated in WP2 and WP3. Small prototype stacks will be tested to demonstrate optimal characteristics.

WP5: PEM FC stack test. The R&D work includes construction and testing of both a LT and HT PEM FC stack using the MEA laminate and bipolar plates developed and fabricated in WP2 and WP3 to demonstrate optimal characteristic

WP6: Pre-pilot line construction. The objectives are to construct a pre-pilot line demonstrating the reproducibility and reliability of the processes on a semi-industrial scale.

WP7: Evaluation of developed fabrication processes. The project will be completed with an evaluation of the technical and commercial feasibility of the developed fabrication routes.

The activities for the project have been divided into two phases. This report concerns the results obtained during the entire project.

The four partners have carried out the work and the main achievements are described below.

Results

WP1. Material & Process Specification

Task 1.1 Performance specification

The performance specifications to be fulfilled by the low cost and high performance MEAs and bipolar plates developed in this project have been define within this task. The specifications as given below are in accordance with the specifications of the Danish PEM Strategy 2008.

<u>DMFC MEA Specifications:</u>

The DMFC MEA specifications targets were set as follow: The maximum cell power electrical output must be up to 0.3 W/cm^2 while the total system power efficiency should be up to 35%. The DMFC MEA's fuel supply is a mixture of methanol and water with an operating temperature lower or equal to 100°C. Regarding the lifetime, the DMFC MEA cells should be capable of reaching over 5000 hours while a DMFC stack should reach over 4000 hours. The target price of a DMFC Stack was 2000 € / kW while a whole DMFC system should come at a price lower or equal to 2500

€/kW. Concerning the DMFC MEA components specification, the Danish Low Temperature PEM Strategy 2008 sets a maximum catalyst loading target at 1 mg Pt/cm^2. The DMFC MEA membrane's conductivity should be at least 0.2 S/cm. These specifications are summarized in the diagram below.

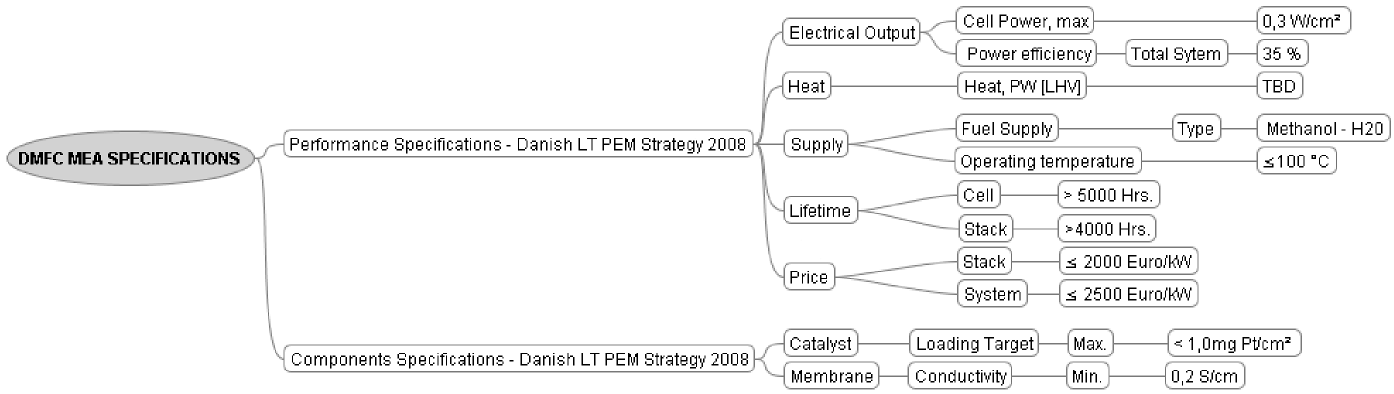


Diagram 1.1.a: DMFC MEA Specifications

Low Temperature H2PEM MEA Specifications:

The H2PEM MEA Specifications targets were according to the Danish Low Temperature PEM Strategy 2008 as follow: Maximum cell power electrical output must be up to 0.6 W/cm^2. The electrical power efficiency should be up to 45% with hydrogen as fuel and 35% with Natural/Reformat gas as fuel. The power efficiency should be 90% with Hydrogen and 85% based on Natural/Reformat gas for the total system. Regarding the lifetime, the H2PEM MEA cell should be capable of reaching over 25,000 hours while a H2PEM stack should reach over 15,000 hours. The target price of a hydrogen PEM Stack is 500 €/kW while a hydrogen based PEM system should come at a target price of 1,000 € / kW and lower or equal to 2000 Euro per kilowatt for a Natural/Reformat Gas based PEM System. Concerning the H2PEM MEA Components Specification, the Danish Low Temperature PEM Strategy 2008 sets a Maximum Catalyst loading target inferior to 0.3mg Pt/cm^2. The H2PEM MEA membrane's conductivity should be at least 0.2 S/cm. These specifications are summarized in the following diagram.

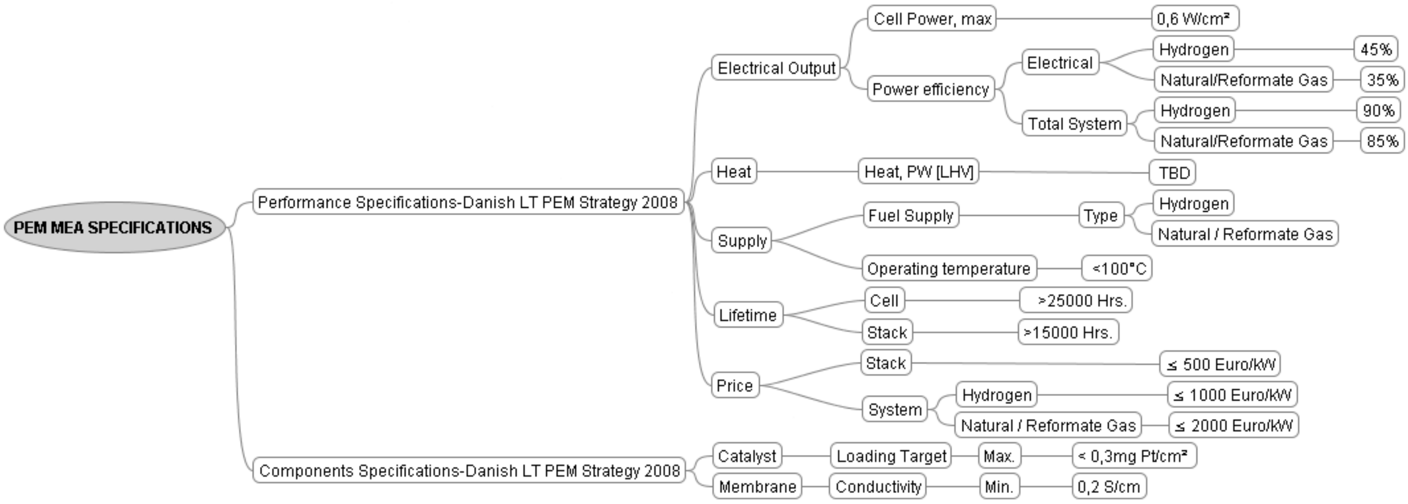


Diagram 1.1.b: Low Temperature H2PEM MEA Specifications

Bipolar Graphite Flowplate Specifications

The flow plates will be operating in environments of Hydrogen, Methanol, steam and air. The process temperature will be up to 200ºC, humidity (RH) 0-100% and PH-value <4. The materials must be stable in both oxidative and reductive environments.
On the mechanical side its necessary with a bending strength of 40 MPa, Compressive strength of 60-80 MPa and E modulus of approximately 12 GPa. These parameters are taken from former projects, where both experimental tests and especially experience in assembling stacks has shown the need of strong mechanical requirements. When assembling a stack, the flow plates are both introduced to bending and compression because of MEA and gaskets.
As flow plates leads the current through the stack, there are high requirement on conductivity. Resistivity or specific resistance [1/σ] of 200 μΩm through plane and 80-100 μΩm in plane must be obtained to ensure the efficiency needed. The permeability of air must be less than $5 \cdot 10^{-5}$ $cm^3/cm^2 \cdot s$.

Task 1.2 MEA laminate process specification

The main technical requirements to be fulfilled by the MEA production line were specified in this task. The requirements were based on experience and state-of-the-art knowledge related to Low Temperature PEM MEA production processes.

CCM Production Line Specifications Based on Spraying Process.

The Production Line was designed for making Catalyst Coated Membrane (CCM) by direct coating of electrode powders onto the polymer electrolyte using a Dry Spraying Process developed by DLR.

The technical requirements for the production line were outlined as follow:

The electrode spraying should be applied on both sides of the membrane at the same time. The spraying should be capable of operating in intermittent mode and with a speed not lower than 1m/minute. The production line should not require more than two spraying nozzles for each side of the membrane. The electrode sizes obtainable should be 10 to 15 cm in length and 8 to 13 cm in width. Maximum electrode loadings to be obtained were 2.0 mg Pt-Ru/cm^2 for the DMFC anode electrode, 1.2 mg Pt/cm^2 for the DMFC cathode electrode, 0.3mg Pt-Ru/cm^2 for the H2PEM Anode and 0.5 mg Pt/cm^2 for the H2PEM cathode electrode.

The control of the applied catalysts amounts should be made possible using a continuous, non-destructive and contact free measuring process.

Task 1.3 Bipolar plate process specification

The main target is to establish cheap and reliable fabrication processes for high quality bipolar plates by either compression molding or by injection molding. The compression molding process is specified and developed in this part of the project. A process layout and specification is listed below for compression molding of a graphite composite containing a thermoset binder.

<u>Preformer</u>:
Preforming of the material is necessary to obtain very accurate molded items and high production rates. The performer handles the material dough ensuring a uniform weight of the bipolar plates.

<u>Press:</u>
The preformed material is compressed into final shape in the bipolar plate mold. The Press and the mold ensure that the main requirement of a thickness variation across the final plate surface is less than 0.05 mm.

<u>Punchout and Grinding:</u>
Punching and grinding of the plates dependent on the mold design will take place in this step removing any superfluous parts.

<u>Oven:</u>
Postbaking might be required to obtain a complete curing, dependent of the binder material used.

An overview of the process layout is shown below (Figure 1.3.a).

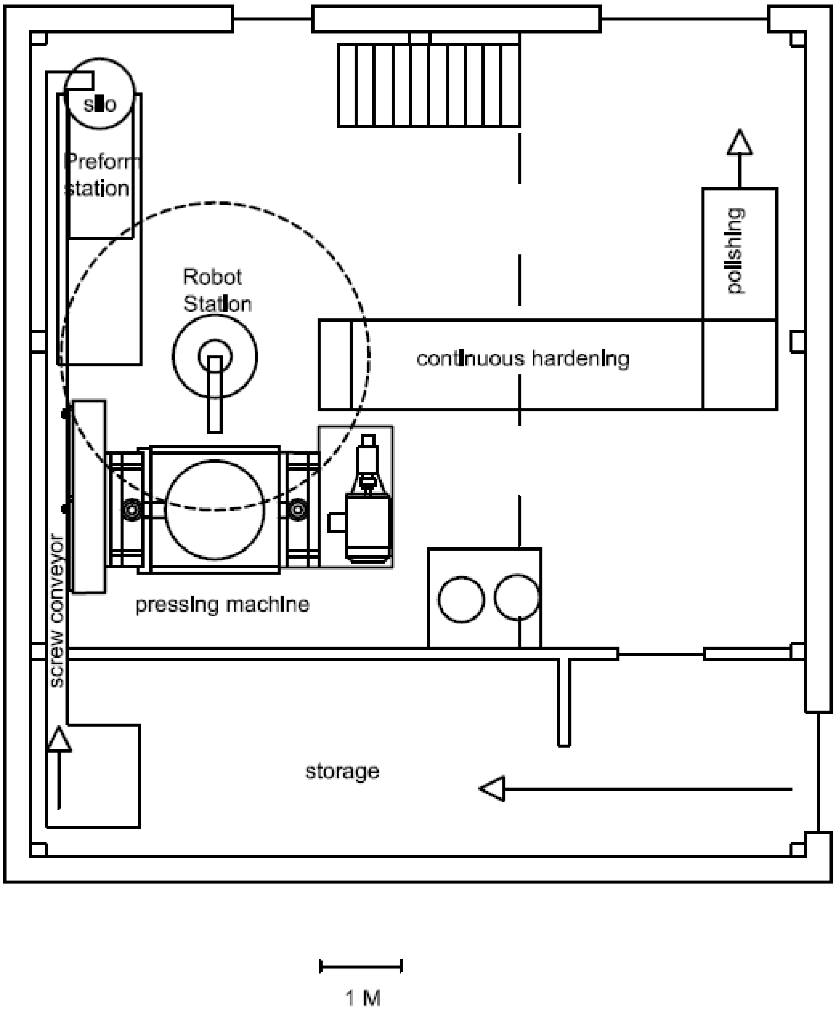


Figure 1.3.a: Process layout for hot pressing flow plates.

The hot pressing flow plate production line was constructed as shown on the layout. The line was tested and verified. The central parts of the line are seen on the figures below.

Figure 1.3.b: The 700 tons hydraulic press with the molds for hot pressing flow plates.

WP2. MEA Laminate Development

Task 2.1 Development of MEA laminate compositions

PEM Laminate compositions including gas diffusion layers (GDL) were defined based on IRD's state-of-the-art performance for low temperature (LT)-PEM MEAs following the current composition of LT-MEA's both for H2PEM and DMFC. The choices of the polymer electrolyte membranes were based on their conductivity, their thickness & their tensile properties. The choice of GDLs was made from their air permeability, their electrical resistivity through plane and their thickness.

Electrode inks which were initially used for wet coating purposes have been developed for the new dry spraying process making two modifications:

- The number of solvents used in the preparation of the inks has been reduced to one thus rendering the ink mixing process easier.

- A unit operation has been added to the ink manufacturing process to allow the transition from wet ink to dry powder while avoiding any alteration of the inks principal components.

Task 2.2 Development of MEA process technology

The MEA production line developed is divided in 3 sections. An overview of the process layout is shown below (Figure 2.2.a) clearly indicating the various steps together with the final layout (figure 2.2.b) including electrostatic filters for filtering any particles in the exhaust air.

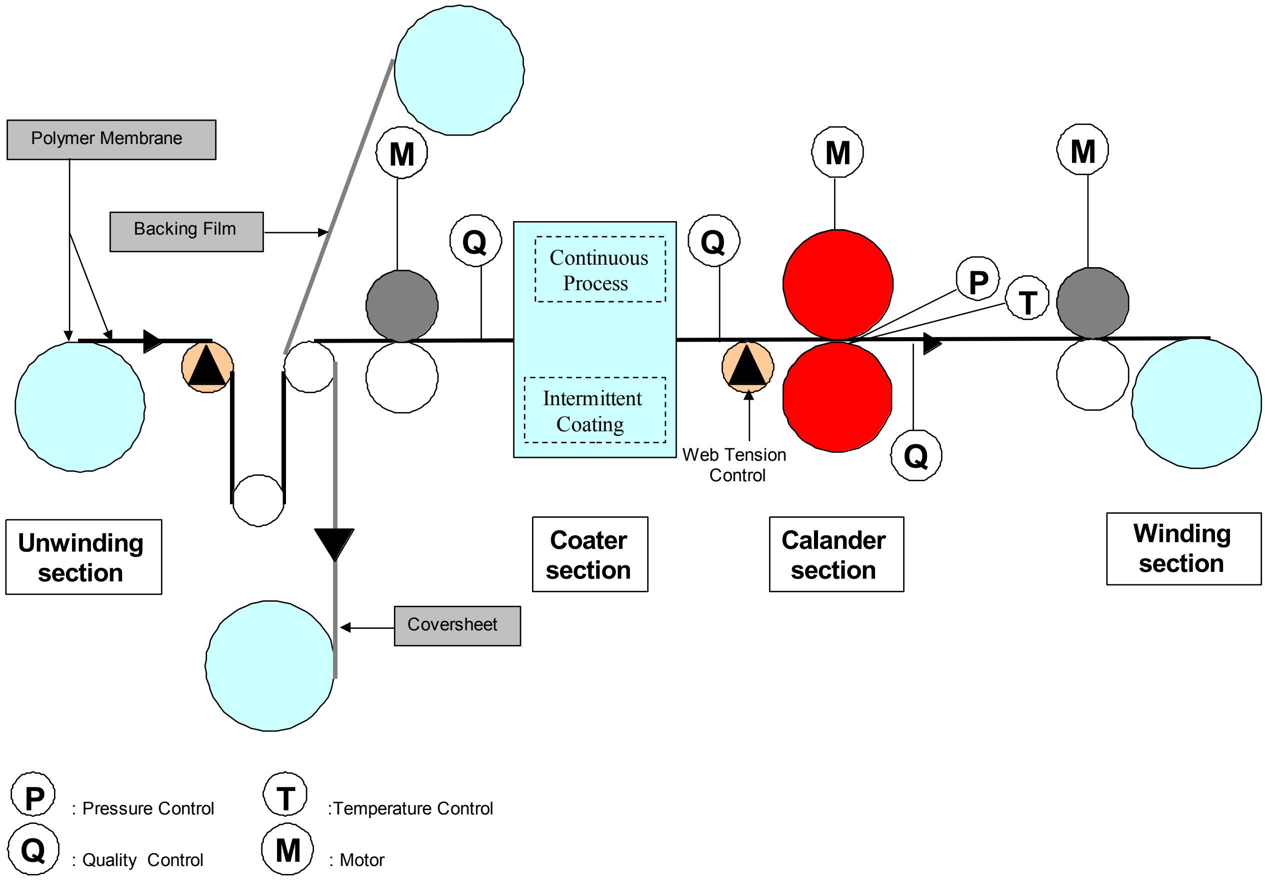


Figure 2.2.a: Layout of Catalyst Coated Membrane Production Line

Unwinding & winding section:
This section allows the use of the polymer electrolyte membrane on roll, state in which they are usually delivered, thus giving the possibility to use the production line in a roll-to-roll configuration and adding a great improvement to the handling of the finished catalyst coated membrane (CCM) prior to its lamination.

Coater section:
This is the section where the electrode powder is transferred onto the polymer electrolyte membrane. Several wet and dry coating techniques were evaluated to achieve such a transfer and it was DLR's state-of-the-art Dry Spraying Process which was chosen to be integrated in the MEA production line as well as state-of-the-art Ultrasonic Spraying Process.

The loadings which have been achieved with the MEA production line are:

- 0,20 – 0,60 mg Pt/cm^2 for PEM Cathode achieved by applying several layers of sprayed Catalyst.
- 0,10 – 0.50 mg Pt-Ru/cm^2 for PEM Anode achieved by applying a layer of sprayed Catalyst.
- 1.00 – 1.80 mg Pt/cm^2 for DMFC Cathode achieved by applying several layers of sprayed Catalyst.
- 1,00 – 2.00 mg Pt-Ru/cm^2 for DMFC Anode achieved by applying several layers of sprayed Catalyst.

The coating technique used involves a certain amount of overspray. The overspray is here defined as the ratio of the amount of electrode powder which doesn't get fixed onto the membrane's surface to the amount of ink powder sprayed towards it. The overspray varies according to the type of electrode powder used. The main trend being a higher overspray for carbon supported catalysts and an overspray range of 20 – 70 %. The overspray materials can be reused or recycled and are as such not wasted.

<u>Calander section:</u>

The rolling and compacting of the CCM increase the contact between the electrode catalyst particles and thereby ensures the performance. The roller is also used to laminate the GDLs to the CCM. The lamination is done at a temperature 20 - 120 ^{O}C.

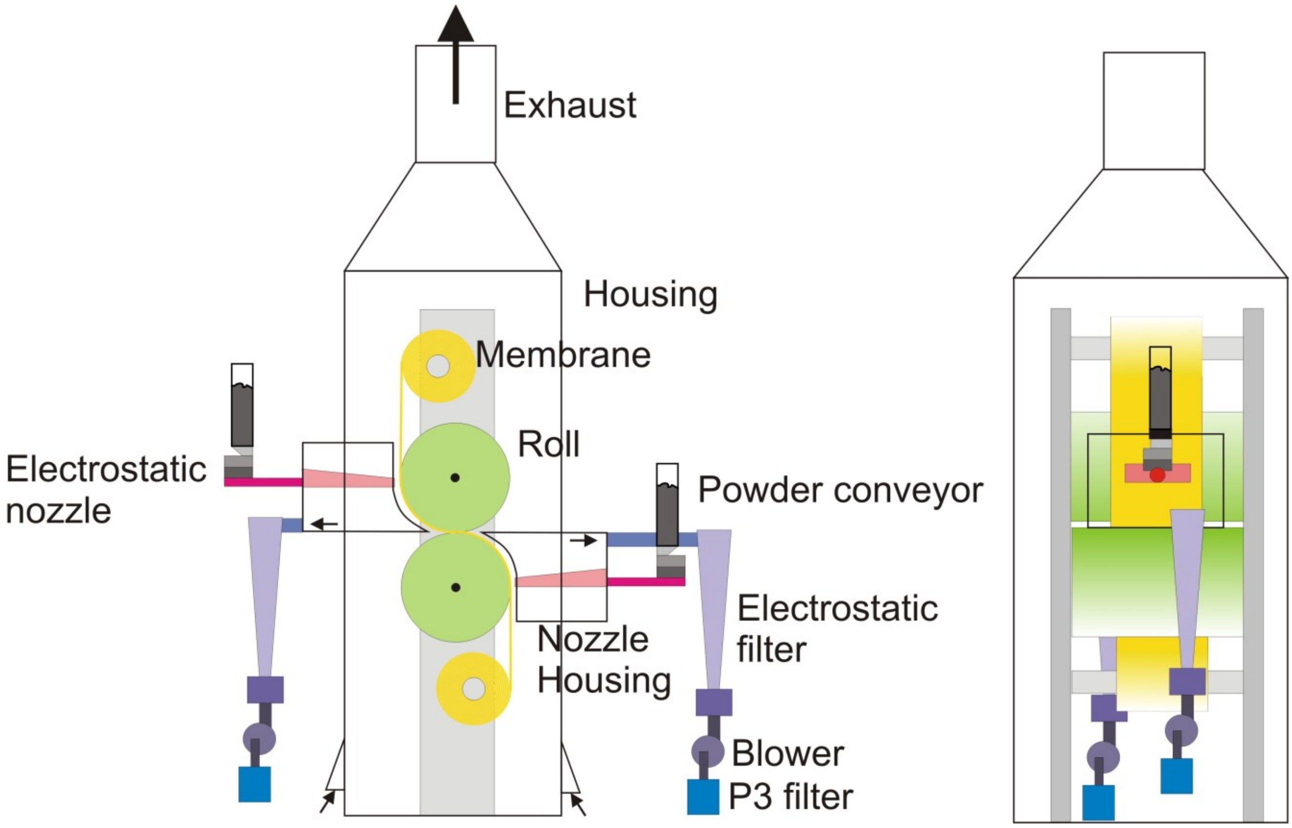


Figure 2.2.b: Final layout of Catalyst Coated Membrane Production Line

The MEA production lines were constructed as shown on the layout. The lines were tested and verified. The central parts of the line are seen on the figures below.

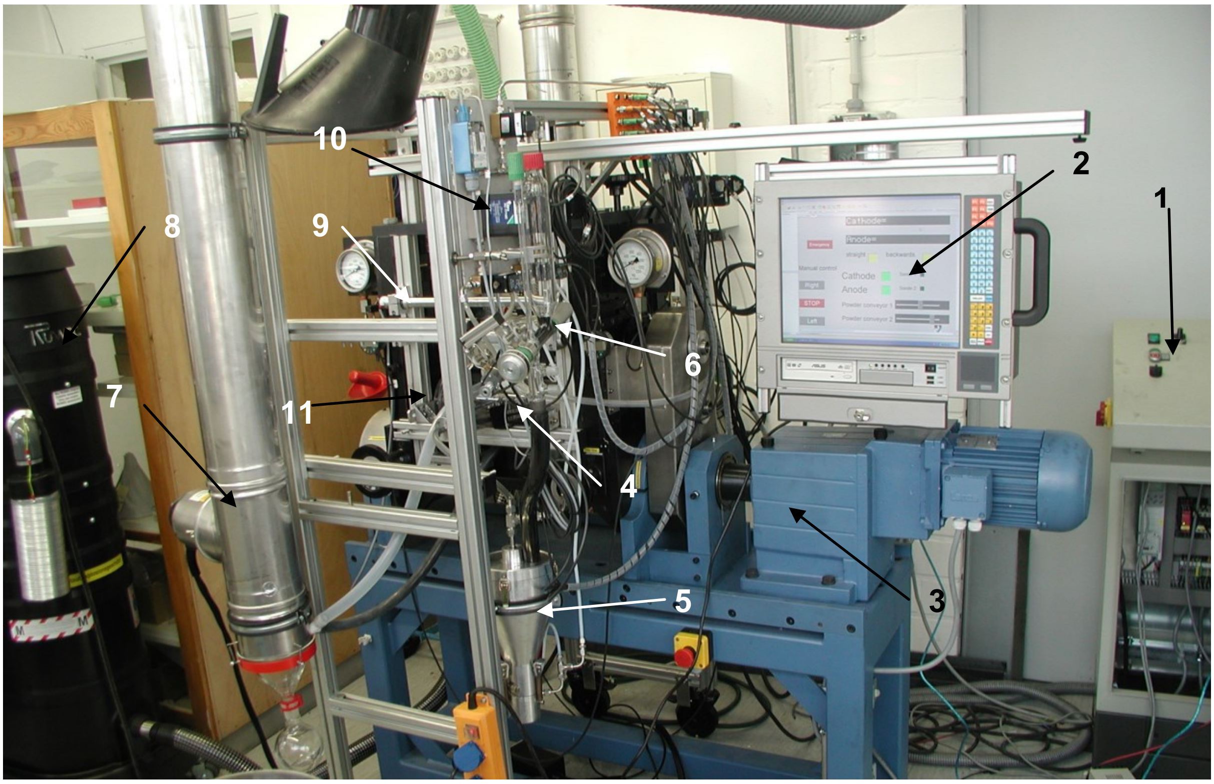


Figure 2.2.c: Catalyst Coated Membrane Production Line including 1. Control box 2. Control panel for manual operation, emergency stop and process information 3. Roll 4. Coating nozzles. 5. Powder conveyors. 6. Powder storages. 7. Electrostatic filters. 8. Vacuum cleaner. 9. Un- and upwind device. 10. Valves and flow controllers. 11. Sensors

Task 2.3 Development of seals

The quest for sealing materials for PEM fuel cells has been divided into two tasks: A low temperature (LT-PEM, 80 deg. C) and a high temperature (HT-PEM, 200 deg. C) application. In both cases the focus is on easy processing by injection molding.

LT-PEM: A thermoplastic elastomer (TPE) capable of withstanding a continuous working temperature of 80 deg. C, resistant to acidic hydrolysis and with a low compression set is sought. A systematic search in electronic databases has been carried out. Most generic materials have been excluded due to the requirement of hydrolytic stability at elevated temperatures. Two grades of newly developed TPE materials have been selected for further testing.

HT-PEM: The search for sealing materials for the high temperature application has been extended beyond thermoplastic materials, and two suppliers of thermosetting elastomers are presently being investigated (Material 2 & 3).

The challenge with sealing HT-PEM MEAs is two-fold:

1. A sealing material should strengthen the PBI membrane around the edge of the electrode.
2. Providing a smooth surface for the sealing of the stack.

The problem is the relatively soft acid doped PBI membrane. This membrane is slightly deformed during the hot pressing of the MEA. The deformation causes stress around the edges of the electrodes, which makes this area prone to crack development. Furthermore, the acid doping of the membrane results in a slight wrinkling of the membrane, meaning that it is difficult to make a tight sealing during stacking. These problems have been reduced by introducing a thin film of PTFE around the electrodes.

DPS have tested several possible solutions to solve these problems. The best results have been obtained by laminating the PBI membrane with a high temperature resistant film. This solution has a number of advantages:

- low uptake of phosphoric acid in film causes less severe operating conditions for the seals used for the stack (i.e. no direct contact with phosphoric acid).
- good adhesion between PBI and the film
- the film material is much stronger than PBI, resulting in much lower failure rate of the MEA's due to crack formation.
- It is possible to obtain a laminate with flat surface, and thereby reduce the problem with leakage significantly.

Chemical analyses

The chemical tests include long term exposure to heat and humidity and methanol, a fuel for the direct methanol fuel cell. Furthermore, extraction tests are being carried out in order to evaluate the dimensional stability of the materials under harsh conditions.

The samples were exposed to solvents, which occur in a fuel cell: a soxhlet extraction revealing the ability to withstand extraction of extractables. The following solvents were used 1: hexane, 2: water, 3: methanol (exposure time: ~1 month at 80 C) 4: 85% phosphoric acid (exposure time: ~1 month at 200 C). The change in sample hardness after extraction is also measured.

Material	Hardness	Process time	Δ mass (%)			
	Shore A	Min.	Hexane	Methanol	Water	Phosphoric acid
Hyflon	70-73	5	+0.1	+0.1	+0.1	-011
Tecnoflon P 959	71	15	+3.3	+3.0	+5.6	+1.1
Tecnoflon - TI modified	58	15	+2.8	+2.7	+4.6	+0.35

Table 2.3.1.: Hardness, process time and extraction data for sealing materials.

These results show that three sealing materials pass the chemical test and the material may well be suitable as sealing material for PEM fuel cells.

Mechanical analyses

To study physical properties of sealing materials and to assess their ability to withstand cyclic thermo-mechanical loading (induced by repeated cooling/heating of FCs), mechanical tests have been conducted in the uniaxial tensile mode. The results have been published in international journals given in the dissemination list below.

The research included (i) tension with various strain rates, (ii) relaxation tests, (iii) creep tests, (iv) cyclic loading with stress- and strain-controlled programs (with various maximum strains, minimum stresses, and numbers of cycles), and (v) fracture tests on DENT (double-end-notched-tensile) samples for evaluation of fracture toughness. All experiments were performed in the entire interval of operating temperatures for PEM FCs.

The objective of the research program was to demonstrate that the sealing materials are suitable for applications, which means that they

- ensure no leakage of gases from FCs during exploitation,
- show a succinctly large hysteresis of energy to adsorb impact loads,
- do not reveal pronounced degradation at elevated temperatures.

Analysis of experimental data reveals that both Hyflon MFA F1530 and Tecnoflon P 959 have good mechanical properties at high temperature. However, none of them were able to fulfil the hardness criteria defined (Shore A between 55 and 60). Therefore, a special version of Tecnoflon with the desired hardness was developed

Process development for sealings
A method for preparation of flat sealing has been developed at Danish Technological Institute. The sealings are made by hot pressing equipment. The results of process development are illustrated in the figures below:

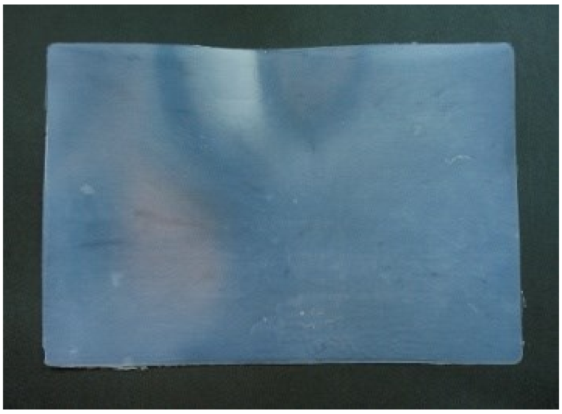


Figure 2.3. a. Flat sealing material of Hyflon MFA F1530

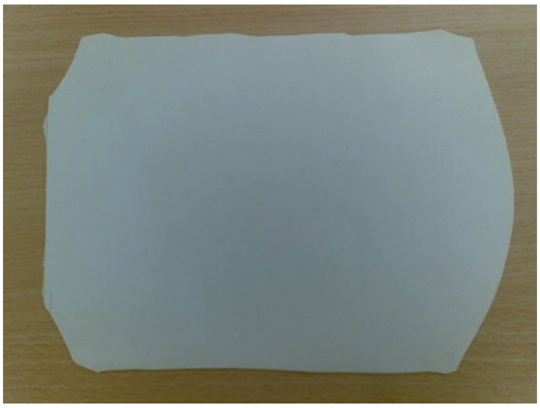


Figure2.3. b. Flat sealing material of TI modified Tecnoflon, having a shore A measured hardness of 58

Also a number of injection molded sealings have been developed in the project. Since the materials demand high temperature processing, a special mould setup has been developed (Figure 2.3.cc) and the processed sealings are illustrated in Figure2.3.e and Figure 2.3.d. The process time for Tecnoflon (TI modified) and Hyflon is 15 min and 5 min, respectively. However, the process time for Tecnoflon may be reduced to between three and five minutes, if the composition is further improved.

Undesired fractures in several of the injection molded Hyflon based sealings were observed. The fractures occur in zones, where two flows of molten polymers meet (Figure 2.3.). The fractures may be avoided by adjustment of mould-design.

Figure 2.3.c. Injection moulding of fuel cell sealings. The mould is developed by IRD. A setup for processing of high temperature polymers has been developed.

Figure 2.3.d. Injection moulded sealing of Hyflon MFA F1530. Process cycle = 5 min.

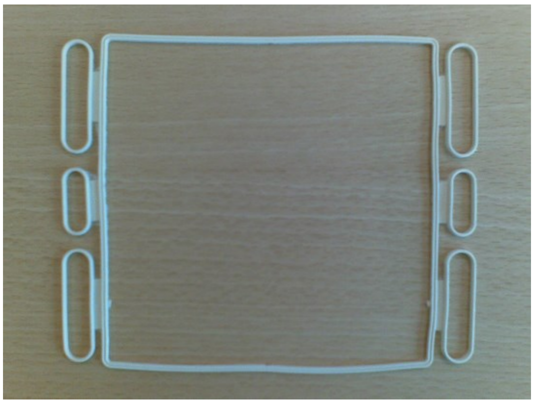


Figure2.3.e. Injection moulded sealing of TI modified Tecnoflon. Shore A = 58.Process cycle = 15 min

Figure 2.3.f. Fracture of injection moulded Hyflon. The fracture may be avoided by adjustment of the mould design.

Both flat sealings and injection molded sealings have been forwarded to IRD and DPS for further processing and have been tested in both low temperature - and high temperature fuel cells with satisfactory results.

Task 2.4 Manufacturing of complete MEA units

MEAs and CCMs were made using the spray coater MEA production lines. The MEAs and CCMs were tested in single cells and stacks in order to verify the production processes.

The MEAs & CCMs produced have been tested using standard single cell test procedures and conditions. The results of the single cell tests are summarized in the graphs below showing the Current-Potential (I-V) performance achievements. For comparison purposes, each one of the graphs also comprises one or several performance results of an IRD Standard MEA. (DMFC MEA 4.00 & 5.00, IRD PEM Standard 601). Results from previous tests achieved under the PORTAPOWER project were also included on the graphs.

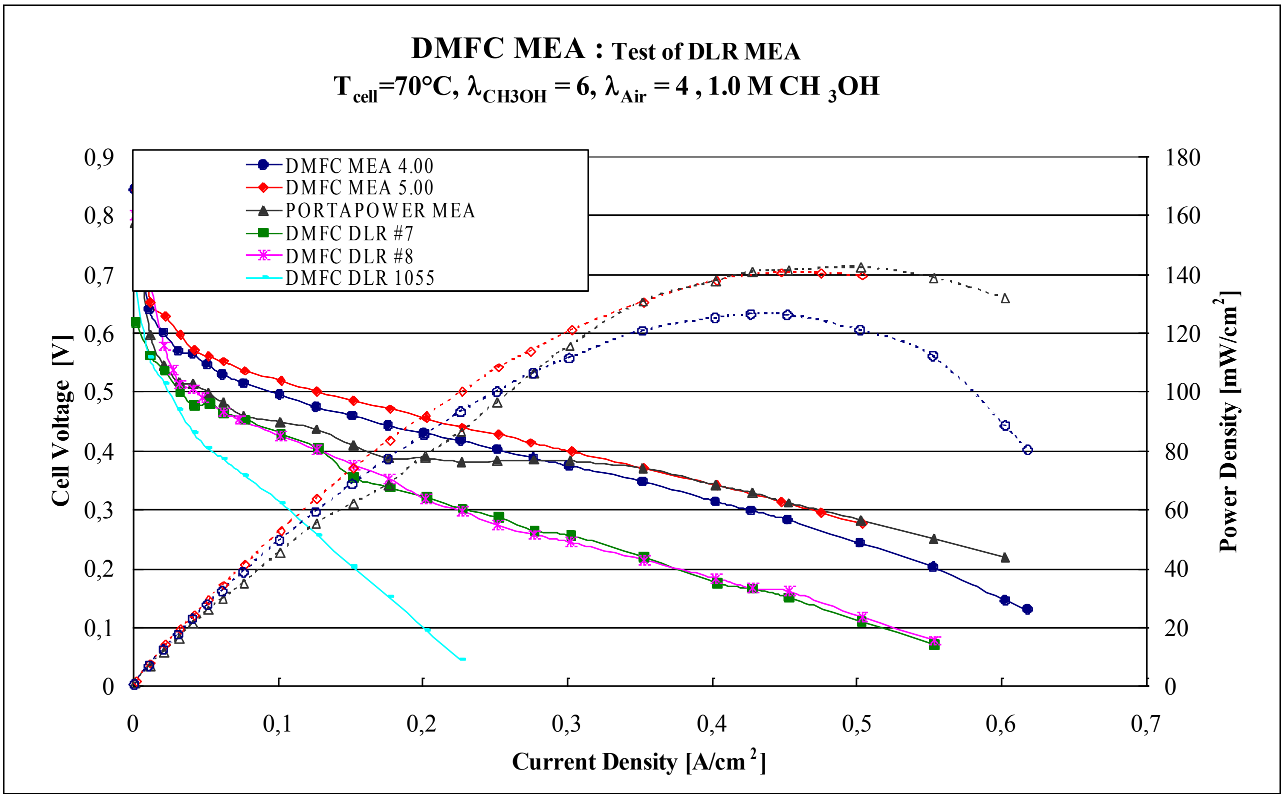


Figure 2.5.a: DMFC MEA Laminate Single Cell IV results.

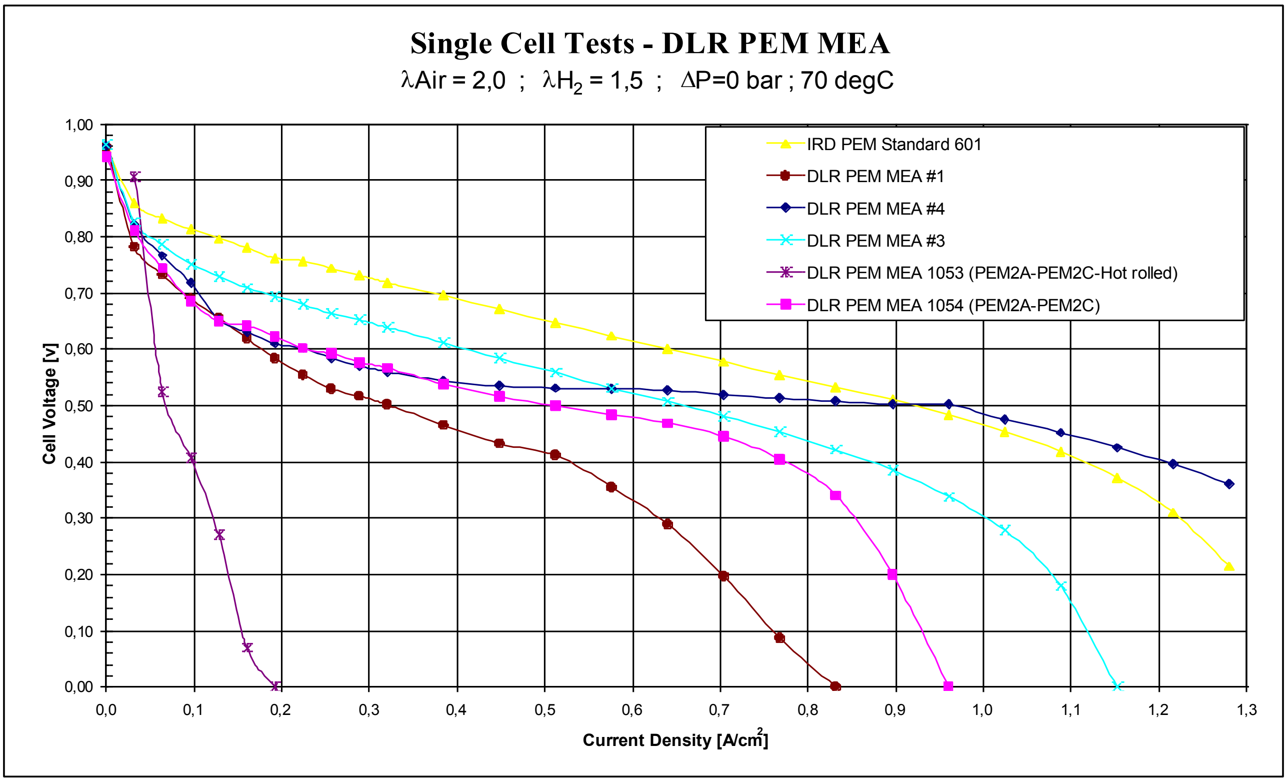


Figure 2.5.b: H2PEM MEA Laminate Single Cell IV results.

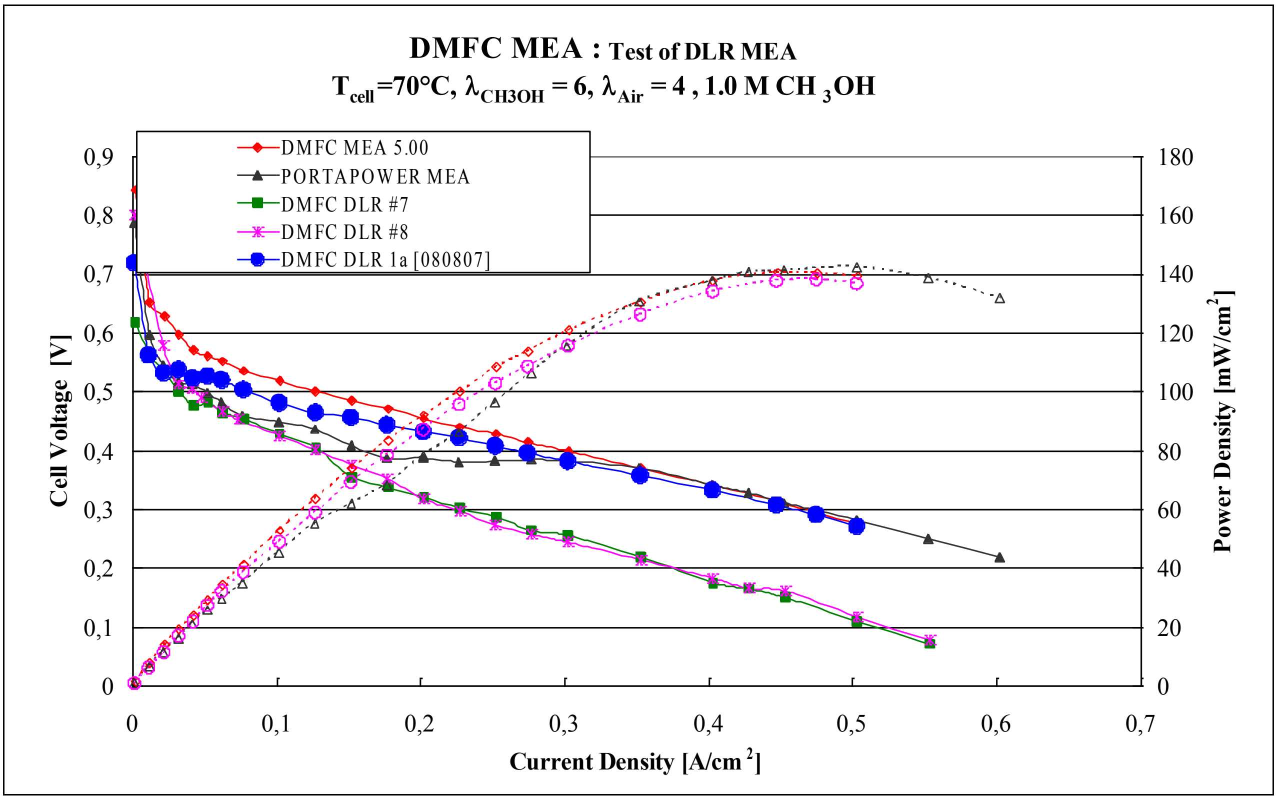


Figure 2.5.c: DMFC MEA Laminate Single Cell IV results.

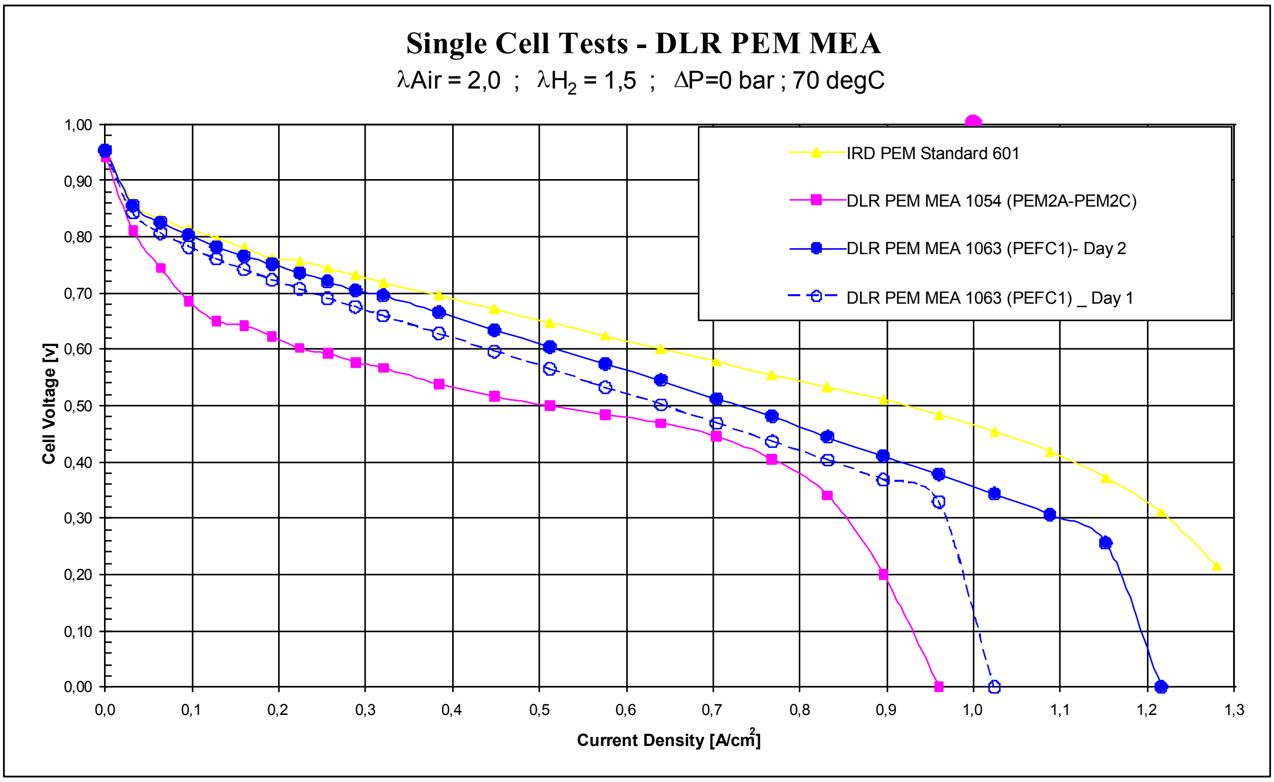


Figure 2.5.d: H2PEM MEA Laminate Single Cell IV results.

The overall conclusion of the single cell tests using MEAs & CCMs manufactured by dry spraying process is that after having encountered some difficulties in the CCM manufacturing process (difficulties which can be visualized by the poor & unusual IV performances of some of the curves on Figure 2.5.a & 2.5.b) it has been possible to solve the encountered problem and new batches of MEAs & CCMs have performed satisfactory as it can be seen on Figure 2.5.c & 2.5.d. All the performance curves on the two last mentioned figures show a usual I-V pattern and they present values only few mV lower than those of IRD's Standard MEA.

WP3. Bipolar Plate Development

Task 3.1 Development of material composition

A systematic choice-of-material based on data for generic types of thermoplastics has been performed. The selection has been done to ensure service temperatures between 80°C and 200 °C under long term exposure together with hydrolytic stability in acidic environments. Two candidate materials for the low temperature application have been identified.

Test moldings have been made with high temperature stable particle filled materials in order to gain some experiences with the mould filling properties of such composite materials. Pictures of the test moldings are shown in figure 3.1.a.

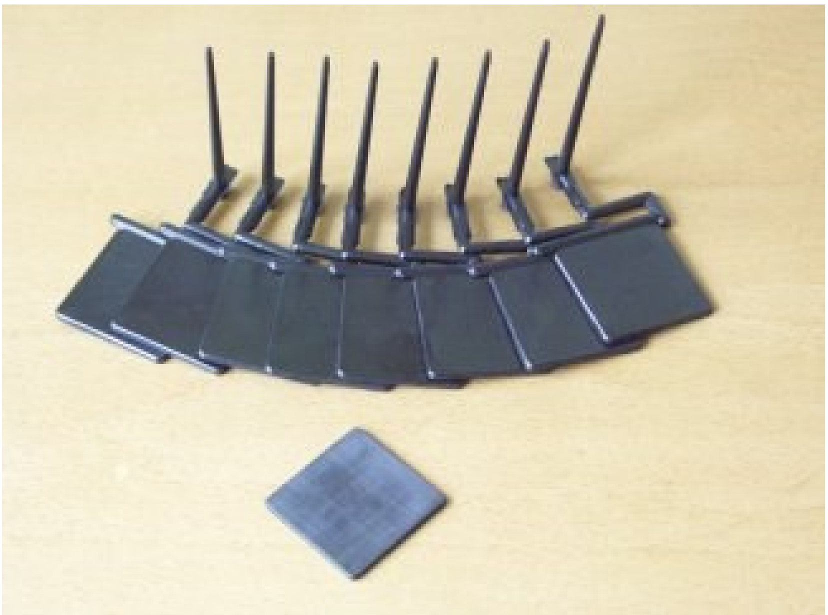


Figure 3.1.a: Test specimens of glass /graphite filled PPS. The simple plate geometry is used as a model for the bipolar plate enabling initial experiments on the molding properties of developed materials.

At Danish Technological Institute focuses have been on injection molding materials using low cost precursors. Mixtures of PE and graphite/CB with sufficient conductivity have been compounded as seen from the electrical measurements. The lowest resistivity has been measured for a PE compound of 14 mΩ·cm, i.e. the selected qualities of graphite and CB are suitable for the purpose, and it is possible to achieve a conductivity which matches the requirements as the resistance is less than 20 mΩ·cm. In the figure below the resistivity of other materials is shown for comparison. (Note the logarithmic scale).

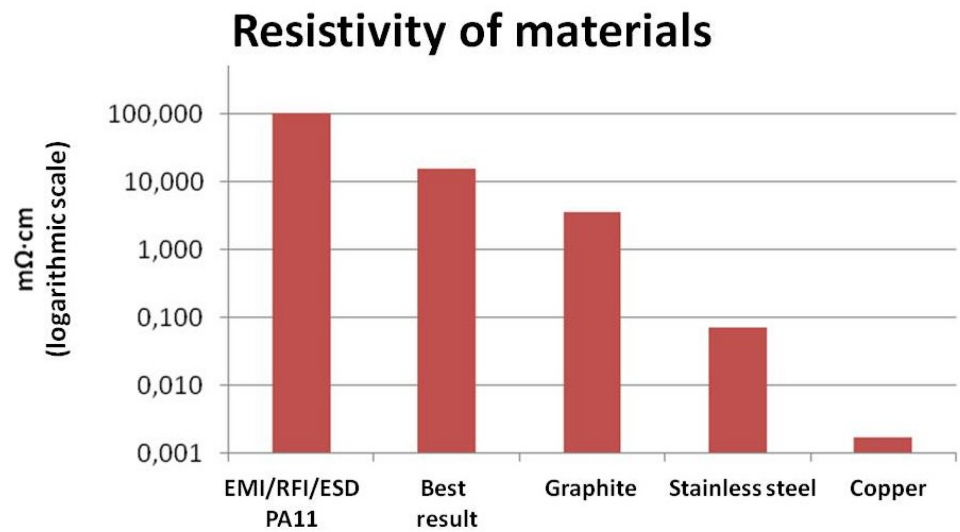


Figure 3.1.b. Conductivity of developed polymer composite in comparison with other conducting materials

Tensile test of conductive PE compounds showed elastic modulus up to 4000-5000 MPa and elongations at break of approx. 1-2% which is insufficient according to the specification. Irradiation tests showed that cross-linking of the polymer in the composites increases strength and ductility. At higher doses, however, a degradation of the polymer took place. As an alternative the use of a moisture curing polyethylene was used as starting material, and compounds were produced and sent to IRD for CNT machining into a test bipolar plate.

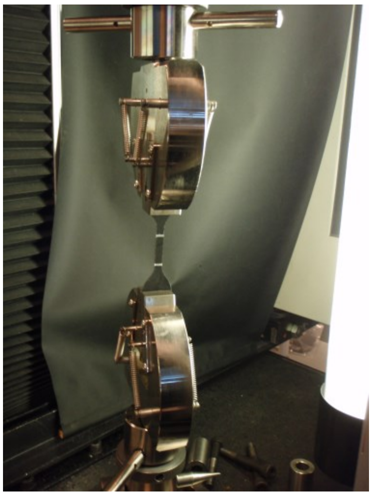


Figure 3.1.c. Determination of tensile strength and elastic modulus of prepared polymer composite

As a high temperature alternative work was done on a PPS based system. A slightly conductive (antistatic/EMI shielding) quality was purchased from Bruno Weile; subsequently, we have tried to increase the conductivity by adding industrial grade multiwalled carbon nanotubes (IG-MWCNT). Although the conductivity of this PPS is several times too low, the experiments with IG-MWCNT showed that it was possible to achieve a doubling of the conductivity by adding only 0.5 wt%.

Further work on composites with IG-MWCNT fillers disclosed that the addition of nanotubes caused the materials to become very brittle, decreasing the elongation at break to very low values. This is thought to be caused by insufficient dispersion of the CNT's in the polymer matrix; a problem that is not unknown in the nanocomposites society. Several other research groups and companies are also working on a solution to this problem, but without success so far.

To measure the conductivity of the developed materials two methods were developed. The most accurate is a four point measurement in-plane; the other is a through-plane measurement. When the flow plate is used in the fuel cell, the current is through plane making this measurement most relevant. Usually, the conductivity is the same in all directions that is not necessarily the case here. The conductive composites are filled with conductive graphite flakes, if the flakes are orientated along the same plane the conduction could be much higher in this plan and in other directions. The graphite flakes could be orientated by the flow making it highly interesting to measure the conductance both different directions in-plane but also though-plane.

With the four point method 4 bands are painted around a long rectangular piece, with conductive silver paint, and the conductance is measured with the usual four point procedure where contact resistance does not influence the result. It is important that the geometry between the two silver bands in the middle is well defined. As a rectangular piece has to be cut from the produced plate, the plate cannot be used for other experiments or used to make experimental flow plates after the measurement.

In the through-plane method, two cobber discs are pressed on a flat sample and the resistance between the cobber discs is measured. The result is thus both the real through plane resistance, but also the contact resistance. To mimic the fuel cell, the material used for the gas diffusion layer on the MEA, was in some measurements placed between the copper discs and the sample. The sample is not damaged by the measurement and can be used afterwards.

Task 3.2: Development of fabrication technologies

Curing of bipolar plates

According to the preliminary tests, no surface treatment is needed to reach the necessary bipolar plate conductivity. If the surface is grinded, it might give better conductivity, but also expose the surface to wear. There will be formed a very thin film enriched with binder material when the flowplate is compression molded. This gives a smooth flow channel. With the selected material and process time currently used, no post baking is necessary for the LT-PEM plates. The post baking process is necessary in molding of HT-PEM plates based on the very temperature stable phenolic resins. The post baking is done for a large number of flow plates at a time. This is a cost effective process.

Preforming of plates

The performing process ensures mixing of raw materials, dosing the materials with a tolerance of +/- 1 g, compresses the preforms.

Compression of flow plates

Pressing flow plates for fuel cells demands a high stiffness of the press construction to obtain a high degree of parallelism. A high pressure, high press speed, and a bump less shift between fast closing of press, and pressing speed are also required. The press specification fulfills these requirements. keeping the maximum deflection of the press daylight area at approx. 60 µm/m.

Task 3.3 Manufacturing of bipolar plates

A range of bipolar flow plates have been manufactured with specified molds in the hot press.

All produced bipolar flow plates have are going through the following QC steps:

- Visual inspection of flow plates.
- Conductivity tests on a number of flow plates produced in a batch.
- Mechanical test on a number of flow plates, dimensions and strength produced in a batch.
- Deflashing of edges and holes.

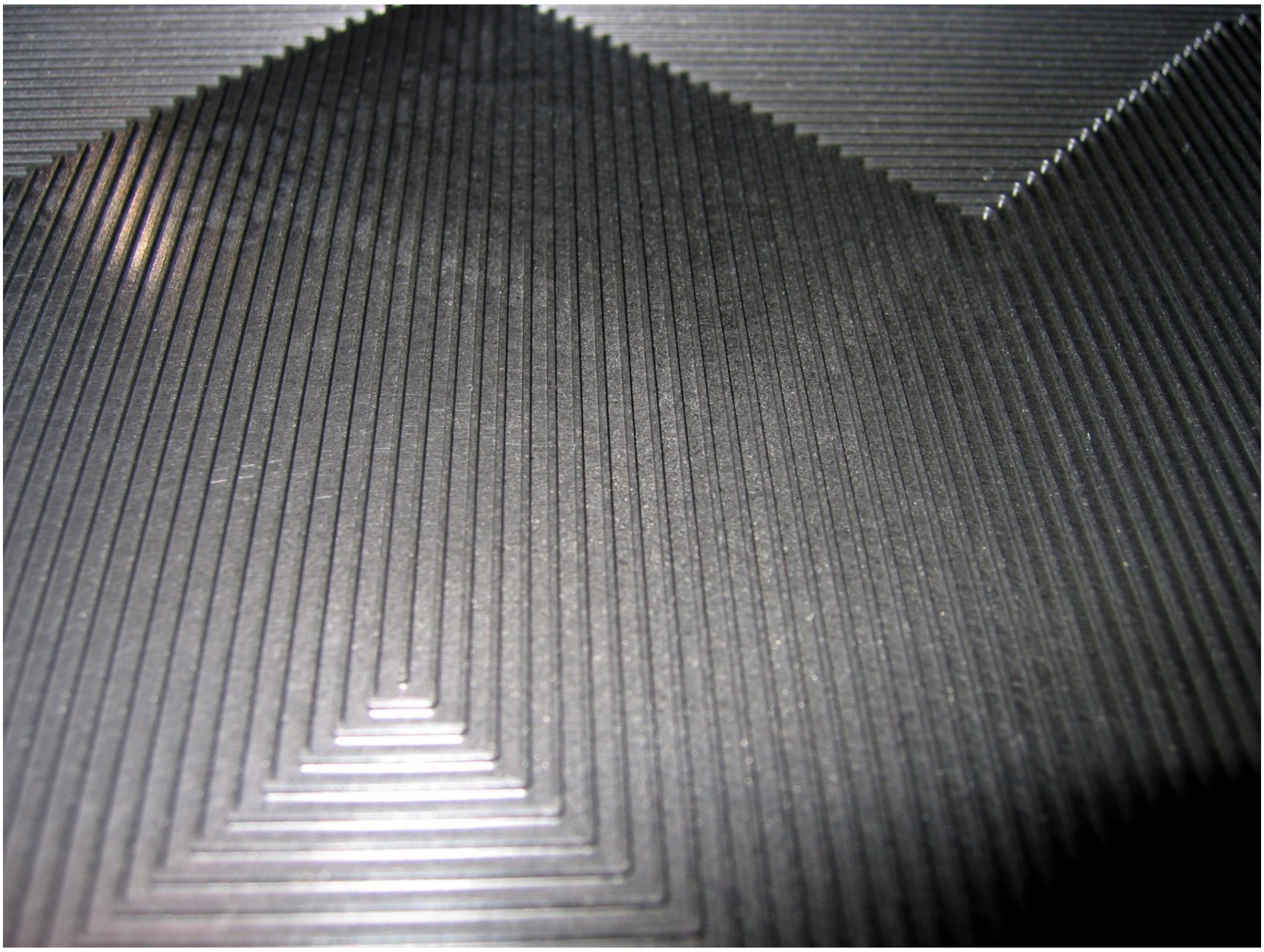


Figure 3.3.a. Molded graphite plate showing the flow field pattern of land and channels ensuring the fuel and air supply to the fuel cells.

The demand for maintenance of the mold cavity is high, since the selected materials have a very low shrinkage factor. The cavity is polished to avoid the molded plates to stick to the surface, but the cavity must be kept very clean for letting the flow plates off the mold.

The properties of the flow plates produces by the developed manufacturing process are reproducible and have a performance that exceeds the required specifications as given in Appendix B.

WP4. Single Cell & Prototype Testing

PEM MEAs were produced on the Catalyst Coated Membrane Production Line and bipolar graphite flow plates molded on the Hot Press Production Line using the material compositions developed. The components were assembled into single cells and stacks for testing of performance and durability verifying the developed production processes.

Task 4.1 MEA test

The PEM MEAs were made with a total loading for both electrodes of 0.8 mg Pt/cm^2 using a 50 µm thick perflourinated polymer electrolyte membrane and 300 µm thick gas diffusion layers with an electrode area of 156 cm^2.

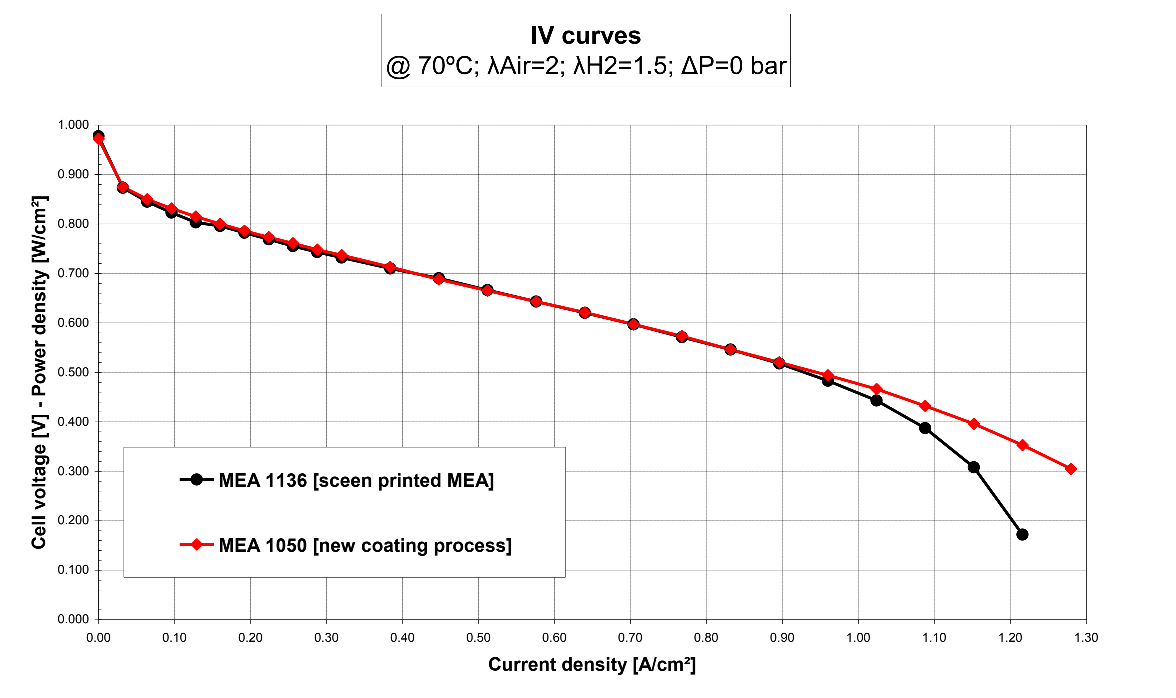


Figure 4.1.a. Performance verification of PEM MEAs produced on the spray coater CCM production line. A MEA made by the previous screen printing process is shown in comparison.

The performance of the PEM MEAs made on the CCM production line compared to PEM MEAs made by the serigraphic process with the same materials and catalysts loadings are slightly better especially at high current density.

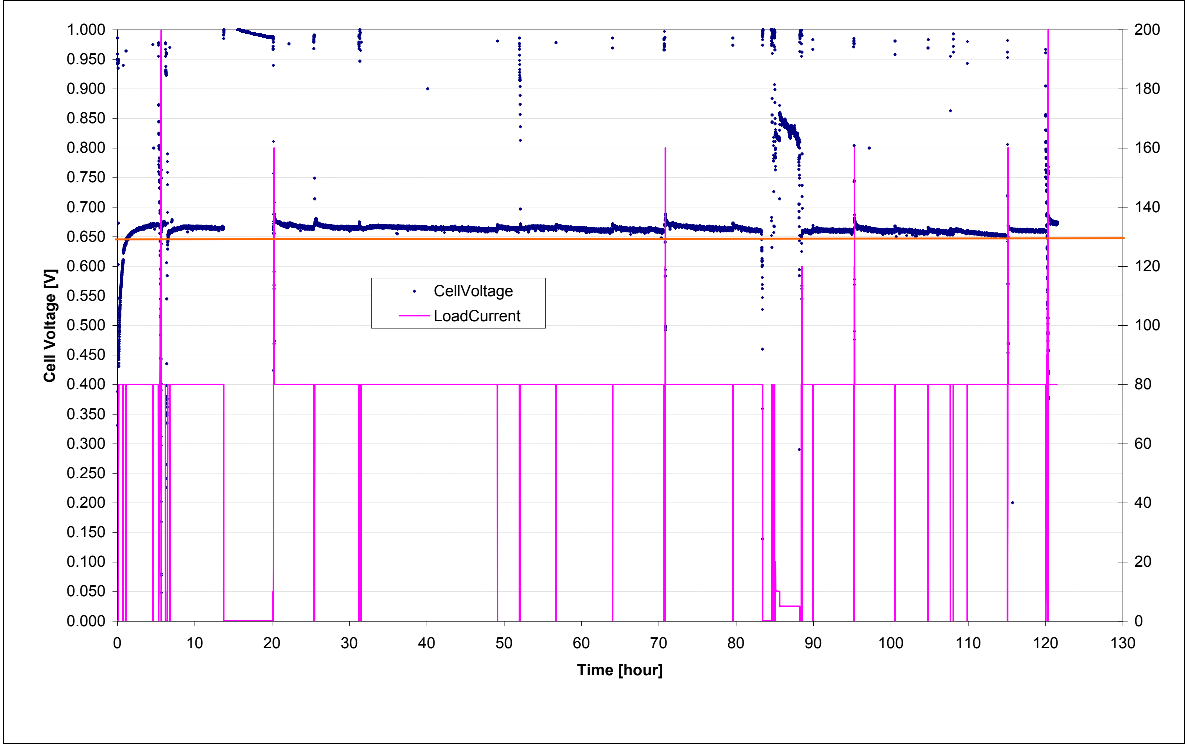


Figure 4.1.b. Verification of PEM MEA durability during a 100 hours test

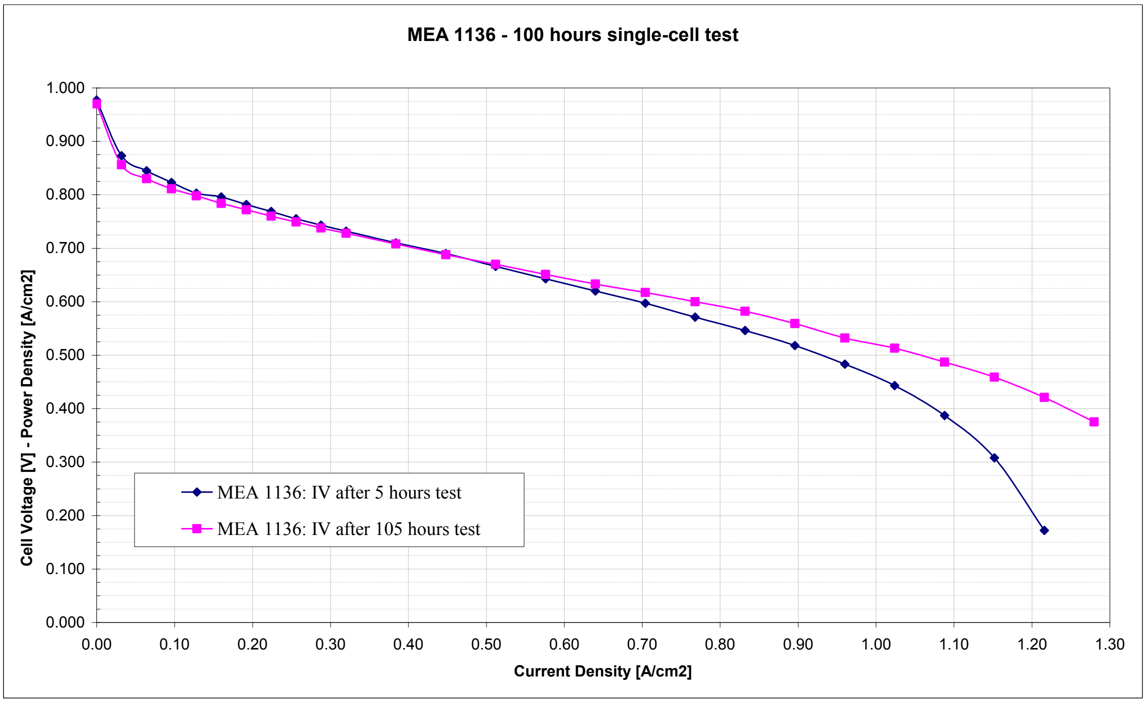


Figure 4.1.c. MEA performance before and after 100 hours durability test.

The 100 hours durability of the PEM MEA shows that a uniform quality of the MEA laminate is produce on the CCM production line. The higher performance obtained after 100 hours test indicate a slow initialization process of the MEA, a phenomenon often seen.

The test results obtained in terms of performance, durability and quality from MEAs produced on the CCM production line verifies the CCM specifications and CCM production process.

Task 4.2 Bipolar plate test

The graphite bipolar flow plates for the PEM and DMFC stacks were molded. For the PEM stacks there were used two plates per cell comprising the anode, cathode and cooling flow fields in a thickness between 2.5 and 3.5 mm. For the DMFC stack only one plate is required since the cooling can be obtained through the fuel circulation. A 35-cell DMFC prototype stack was manufactured and assembled. The obtained performance compared to the specified performance characteristics is shown if Figure 4.2.a. The test results plotted showed that the performance more than fulfilled the targets verifying the plate specification and plate fabrication process.

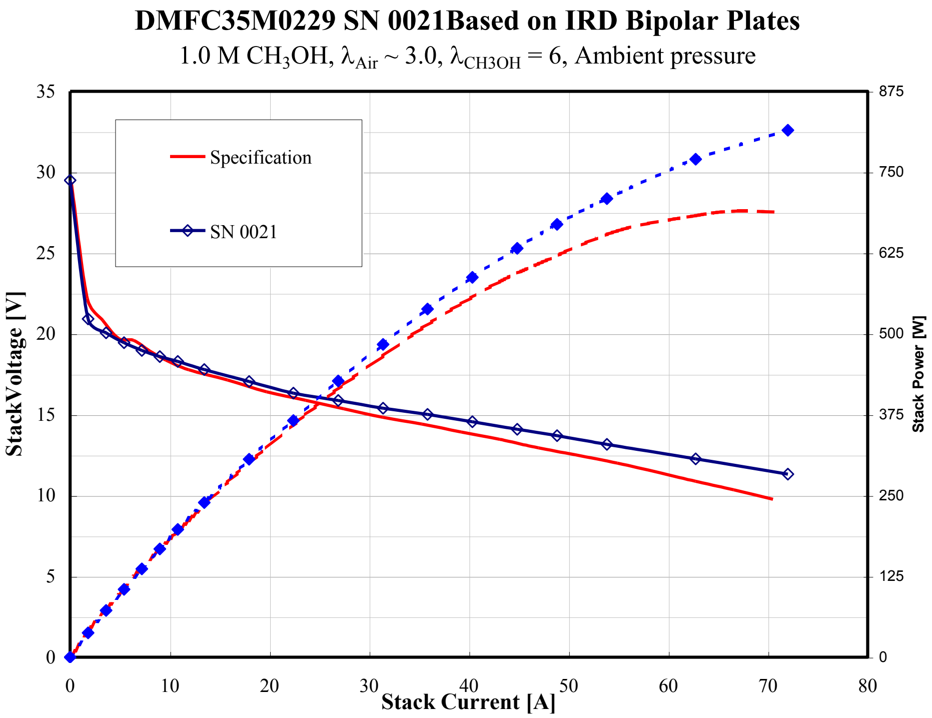


Figure 4.2.a. Performance of prototype DMFC stack

Based on these positive results, three 1 kW stacks aimed for demonstration in the project "EFP-06 DMFC MEA and Stack Development" (J.No. 33032-0145) were fabricated using the developed manufacturing process lines. The results are shown in Figure 4.1.b, all fulfilling the target performance demonstrating the reproducibility of the production process.

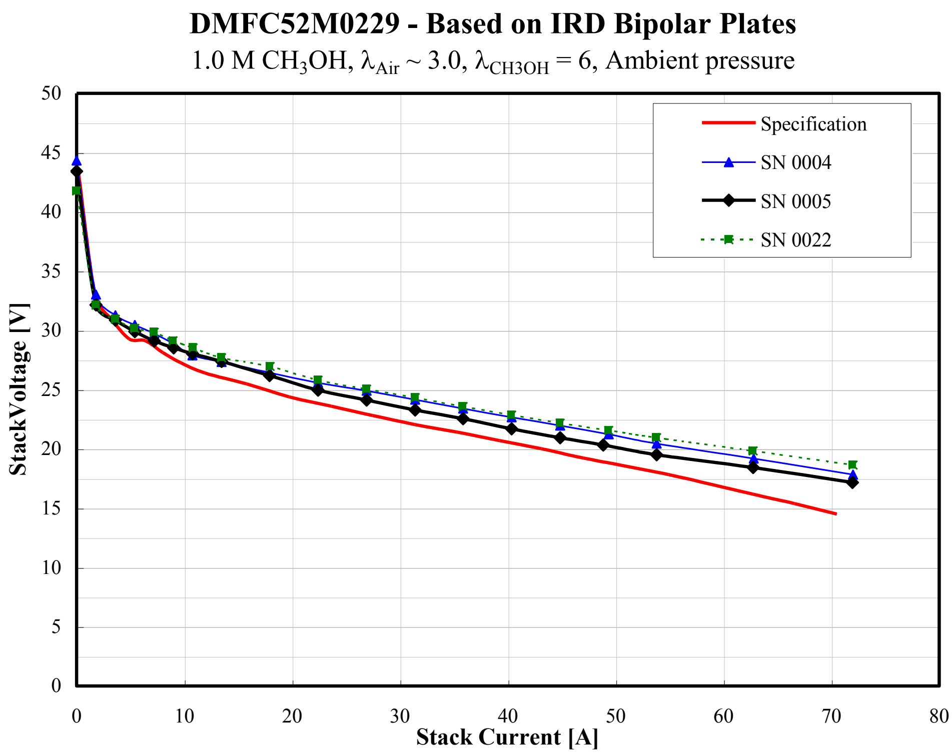


Figure 4.2.b. Performance of DMFC stacks to be used in demonstration project.

WP5. PEM FC stack test

Task 5.1. Test of LT-PEM FC stacks

Production runs of PEM MEAs and graphite bipolar plates were done on the CCM production line and the hot press flow plate production line. The components were tested in sections of complete stacks in order to establish the reproducibility of the production processes and the durability of the MEAs and plates.

The results of these PEM stack tests are shown in Figure 5.1.a and Figure 5.1.b

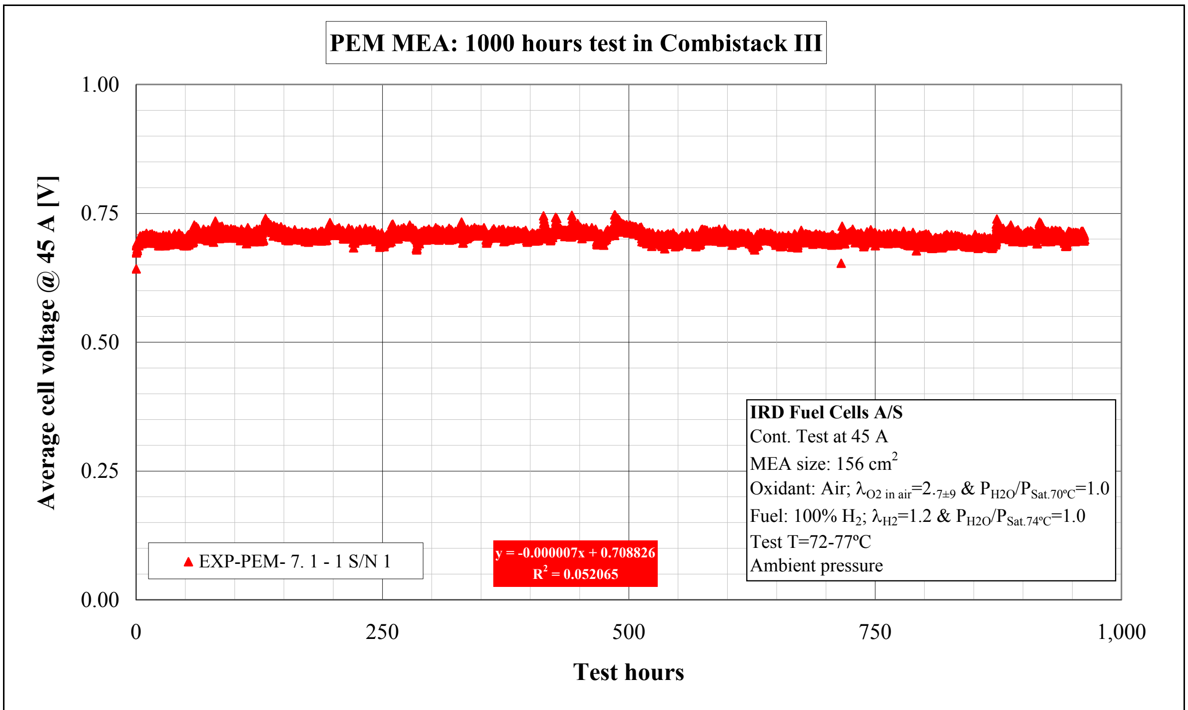


Figure 5.1.a: 1000 hours test showing average cell voltage of MEAs made on the CCM production line.

The initial average cell performance in this section of the PEM stack was 709 mV @ 45 A degrading to an average cell voltage of 702 mV @ 45 A after 1.000 hours test at a constant load of 0.3 A/cm^2 (45 A). The observed degradation rate was 7 µV/hr corresponding to an estimated lifetime of 15.000 hours (15% performance reduction).

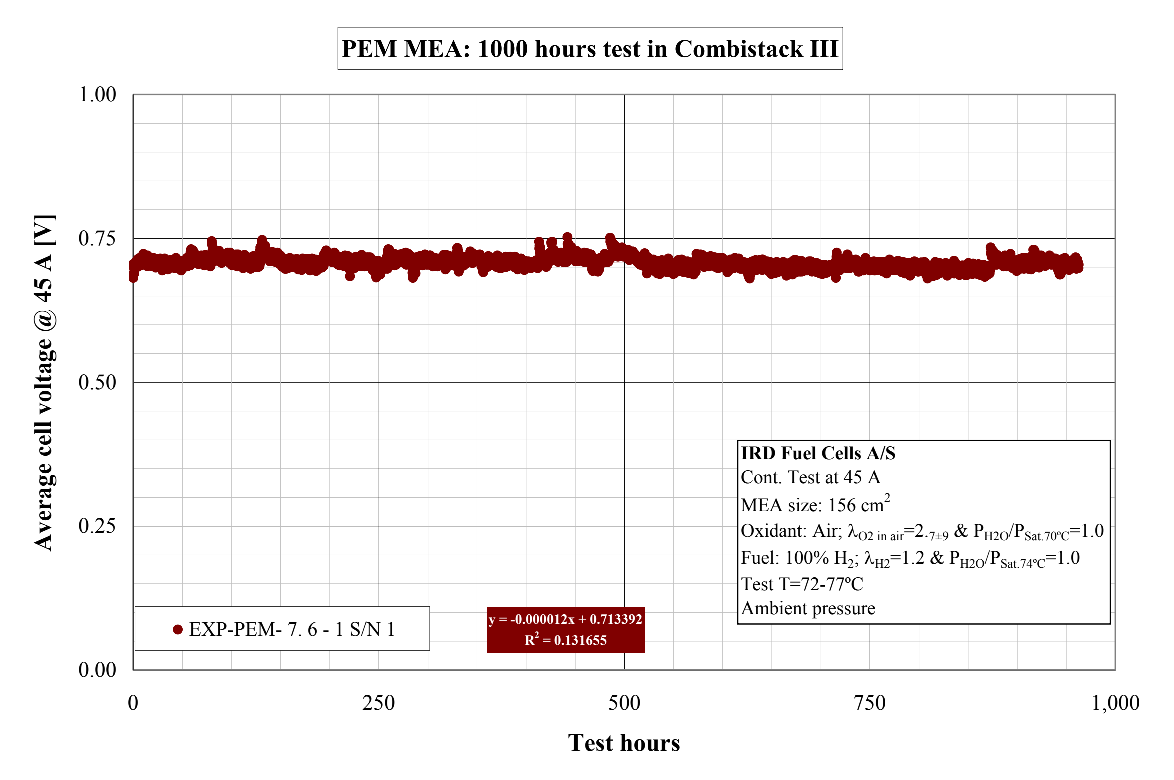


Figure 5.1.b: 1000 hours test showing average cell voltage of MEAs made on the CCM production line.

The initial average cell performance in this other section of the PEM stack was 713 mV @ 45 A degrading to an average cell voltage of 702 mV @ 45 A after 1.000 hours test at a constant load of 0.3 A/cm^2 (45 A). The observed degradation rate was 7 µV/hr corresponding to an estimated lifetime of 10.000 hours (15% performance reduction).

The results show that the production lines have sufficient reproducibility. The life time expectations obtained through the tests are in accordance with the project targets. The MEA performance and specifications are given in Appendix A.

Task 5.2. Test of LT-DMFC stacks.

Production runs of DMFC MEAs and graphite bipolar plates were done on the CCM production line and the hot press plate production line. The DMFC MEAs and the plates were assembled into stacks and at the same time a DMFC stack was assembled with commercial ref. bipolar plates Durability tests of DMFC stacks based on IRD LT0 bipolar plates and on commercial ref. bipolar plates respectively are shown in, Figure 5.2.a and 5.2.b

The stack based on commercial ref. bipolar plates has been running for 2750 hours and the stack based on IRD LT0 for 3250 hours. The stacks are very similar in behavior and the estimated lifetime using EoL defined as 0.33 V @ 70 mW/cm^2 are for both stacks 5.000 hours (Figure 5.2.c).

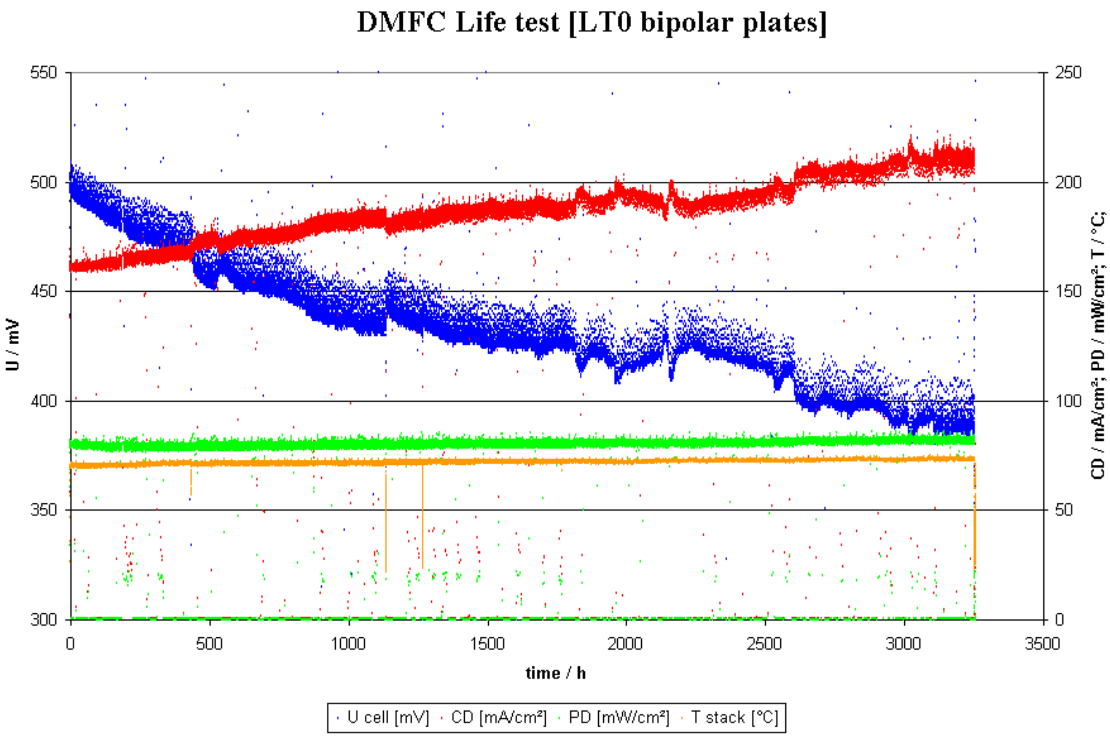


Figure 5.2.a. Life test of DMFC stacks with IRD graphite composite bipolar plate. Tested at constant power.

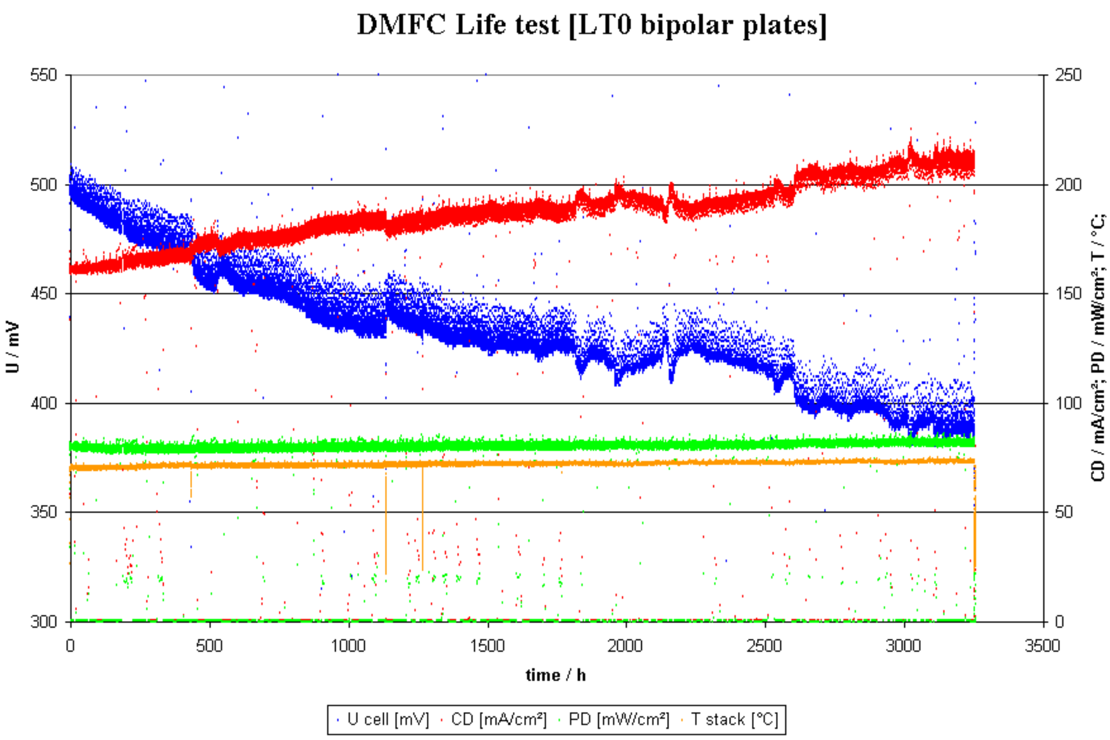


Figure 5.2.b. Life test DMFC stack with commercial ref. bipolar plates. Tested at constant power.

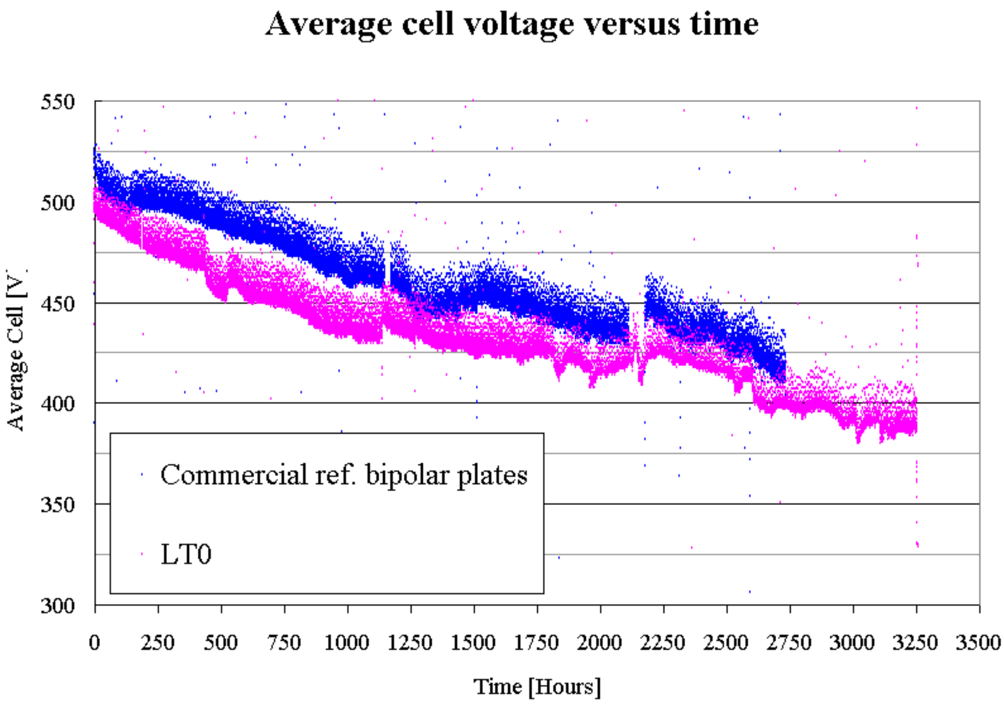


Figure 5.2.c. DMFC life test comparing stacks with different bipolar plates.

Task 5.3 Test of HT-PEM FC

Blank bipolar graphite plates thermally stable up to at least 180 ^{o}C have been produced on the hot press production line. Flow fields have been machined. CCM spray coating processes have been demonstrated with the HT-PEM formulation. The results obtained have not been reproducible, but by development it is believed that the CCM production line also will be capable of producing HT-PEM MEAs. The HT graphite plate fulfils the specifications and is being produced and used in other HT-PEM projects.

WP6. Evaluation of the fabrication routes developed

The technical and commercial feasibility of the developed fabrication routes have been analyzed. The specifications in WP1 have been followed in this work package by a detailed cost analysis to pinpoint cost-driving materials, processing routes, and components during the development work.

Task 6.1 Cost benefit analysis for flow plates

Flowplate price elements

Material per flowplate will initially be 1 € for low temperature flow plates. This is based on current material prices. The cavity share will depend on the number of plates made but typically be approx. 0.25 – 0.50 €. All together the target of 1.5 € pc. or 50 €/m^2 is fulfilled.

<u>Current Flow plate price.</u>
Machined flow plates costs above 50 € pc. The blank flowplate is bought from sub supplier costing 20 -30 € pc.

<u>Cost reduction on complete stack.</u>
The 1 kW DMFC stack contains approx. 50 identical flow plates.
This means that the flowplate share in price is reduced from 2666 € to 75 €. A remarkable change in cost price which increase the commercial potential.

Task 6.2: Evaluation of the technology

Materials and processes have been developed which are allows accurate, reproducible and cost effective production of both MEAs and bipolar graphite flow plates.

The technical specifications obtained for the PEM and DMFC MEAs produced by the developed processes are compared in the table below with the targets stated in "Strategiplan for PEM" and in the road maps for the particular technology[1)]:

PEM MEA	**Status (2010)**	**2012**	**2020**
Ledningsevne	0,2 S/cm	0,2 S/cm	0,3 S/cm
Levetid	10.000 h	12.000 h	50.000 h
Effekttæthed @ 0.7 V	0,3 W/cm^2	0,3 W/cm^2	0,7 W/cm^2
Effekttæthed Max	0,5 W/cm^2	0,65 W/cm^2	0,9 W/cm^2
Platinforbrug/elektrode	0,4 mg/cm^2	< 0,3 mg/cm^2	<0,2 mg/cm^2

DMFC MEA	**Status (2010)**	**2012**	**2020**
Ledningsevne	0,2 S/cm	0,2 S/cm	0,3 S/cm
Levetid	3.000 h	5.000 h	10.000 h
Effekttæthed @ 0.4V	0.10 W/cm^2	0,13 W/cm^2	0,35 W/cm^2
Effekttæthed @ Max.	0.12 W/cm^2	0,18 W/cm^2	0,40 W/cm^2
Platinindhold/elektrode	1,5 mg/cm^2	<1,5 mg/cm^2	<0,5 mg/cm^2

The status for the PEM MEA for 2010 compares well with the targets for 2012. The lifetime must be improved together with the max. power density and a reduction in the platinum loading.

The status for the DMFC MEA for 2010 compares also well with the targets for 2012 requiring and longer lifetime which the results presented for WP5 also indicates will be reached and a lower platinum content.

The stack prices for the LT PEM and DMFC stacks based on the MEA laminate and bipolar plate production cost have been calculated. The results are given in the table below and compared with the national Danish targets as stated in the "Strategiplan for PEM" and the road maps for the particular technology [1)]:

Year	LT PEM Stack (€/kW)	DMFC Stack (€/kW)	HT PEM Stack (€/kW)
Status	5.200	13,000	
2008	20,000	15,000	15,000
2012	3.100	9,000	3,000
2020	1.200	1,500	400

[1)] targets defined in the national Danish road maps on PEM FCs, latest revision April 2011

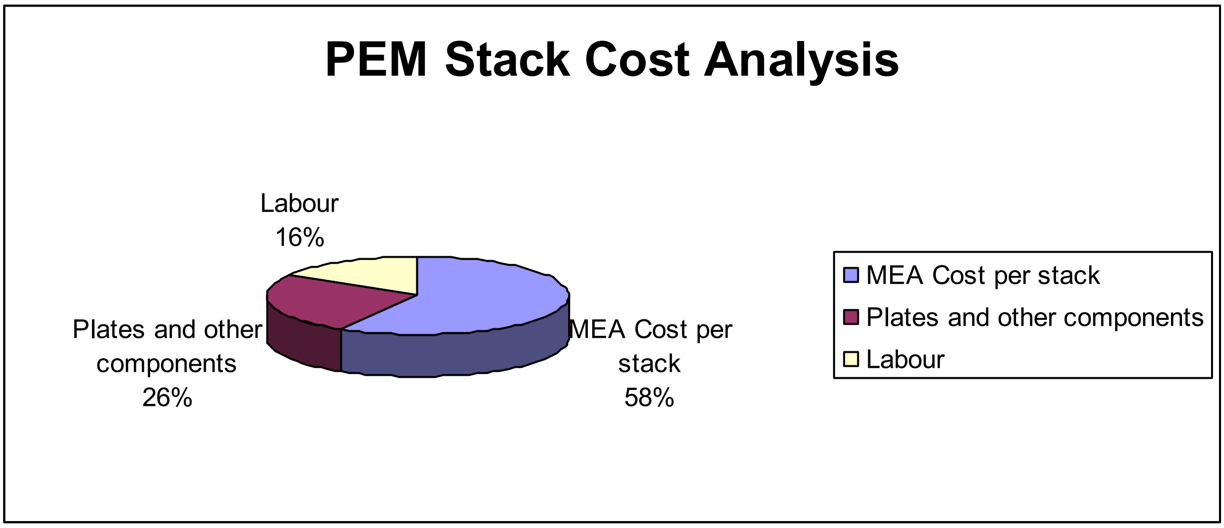


Figure 6.2.a. The PEM stack cost distributed between different categories.

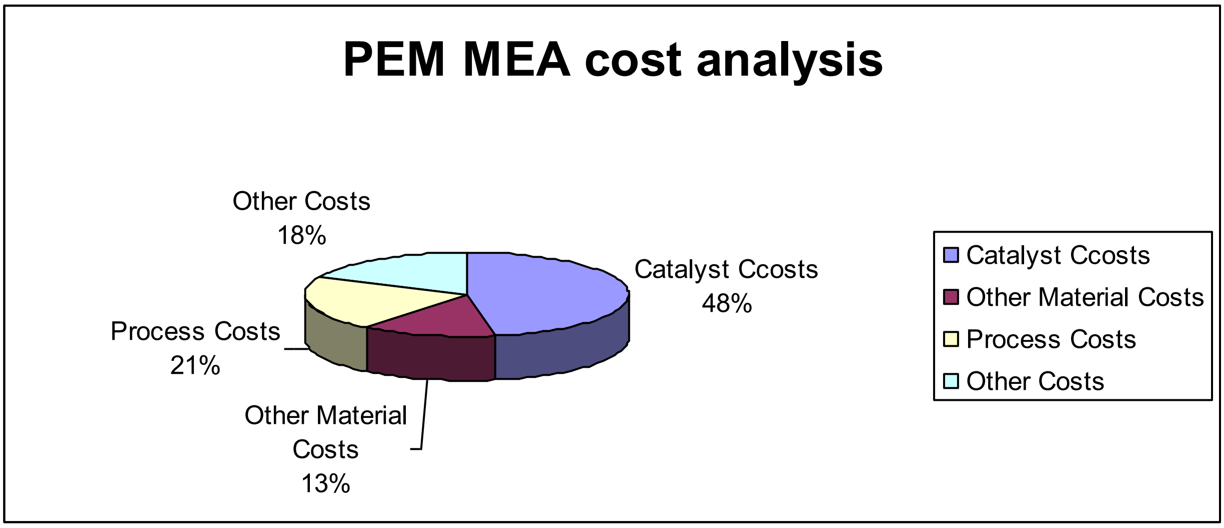


Figure 6.2.b. The PEM MEA cost distributed between different categories.

The PEM and DMFC stack prices are reasonably in accordance with the target values given in the table above. An analysis of the stack price structure (Figure 6.2.a and Figure 6.2.b) shows that the MEA catalyst cost together with production costs are the dominating price factor. Research and development effort in order to reduce the prices within the PEM fuel cell technology should be focused on reducing the amount of catalyst used in PEM Fuel Cell and reduce production costs

Conclusion

The following have been achieved during the “Development of Low Cost and High performance MEA - and bipolar plate processes” project:

The MEA laminate process and production line, the bipolar plate process and production line together with the sealing materials have been finalized and verified.

The MEAs manufactured meet the performance requirements.

The bipolar plates manufactured meet the performance and cost requirements.

The project has in overall terms achieved the technical milestones.

ANNEX A: MEA specifications for commercial sales

MEAs for Fuel Cells

(Membrane Electrode Assemblies)

MEA Solutions for PEMFC and DMFC Products

MEAs are the key component determining fuel cell performance and efficiency. IRD employs a team of superior MEA specialists, and can provide high performance Catalyst Coated Membranes (CCM) and MEA structures, based on Cabot's catalysts, for a range of applications.

IRD PEM MEA PERFORMANCE:

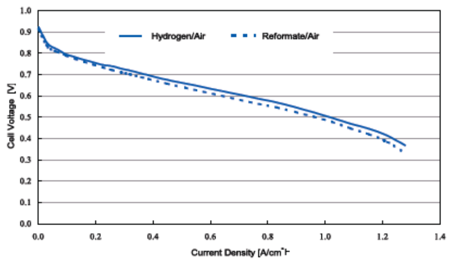
Ambient pressure
Temperature: 70°C
Cathode: $\lambda = 2.0$
Anode: $\lambda = 1.2$
Humidification: PH2O/PSat.70°C = 1.0
Reformate: 75% H_2, 25% CO_2, 50 ppm CO/2% Air bleed

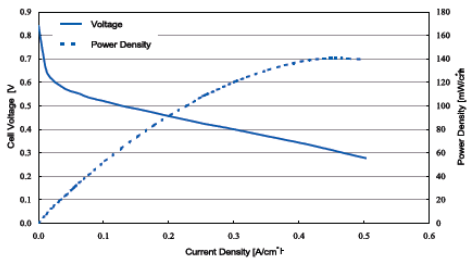
DMFC MEA PERFORMANCE:

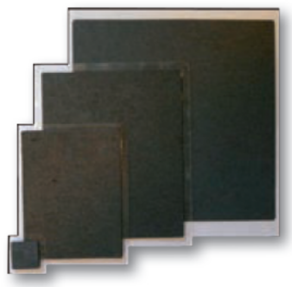
Ambient pressure
Temperature: 70°C
Cathode: $\lambda = 3.0$
Anode: $\lambda = 4.0$
Fuel: 1.0 M CH_3OH

IRD

IRD Fuel Cell Technology has been working with R&D, design and manufacturing of Fuel Cells since 1982, and is today among the leading experts in fuel cell technology. IRD Fuel Cell Technology has recently acquired Cabot Corporation's MEA manufacturing plant in Albuquerque, New Mexico. IRD and Cabot have entered into joint development and supply agreements, collaborating closely on the development of improved MEAs. By incorporating Cabot's advanced electrocatalysts, the performance and superior quality of IRD's MEA products and new non-Nafion solutions are increased even further.







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For additional information, please visit our website at www.ird.dk, or contact:
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ANNEX B: Flow Plate specifications for commercial sales

Flow Plates for Fuel Cells

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Manufacturing technology:
IRD can offer low cost press moldings for high volume serial production, CNC machining for small series prototype plates or blank composite plates for your own processing tooling.

Flow channel design
IRD can manufacture flow plates in all sizes with your proprietary pattern design. You may also purchase one of our existing and well proven designs, or let us help you to design a new pattern specifically for your application.

Product Advantages
IRD Flow Plates offer excellent electrical and thermal conductivity at competitive prices, using state of the art flow plate design combined with carefully selected raw material compounds and mass production techniques.

IRD Flow Plates Technical Properties

Physical Properties

Bulk Density	1.83 g/cm³
Water Adsorption	‹ 0.1 %

Mechanical Properties

Flexural Strength	43 MPa
Compressive strength	76 MPa
Young Modulus	10 GPa

Electrical properties
Specific electrical resistance

in plane xy	100 μΩm
through plane z	200 μΩm

Thermal Properties

In plane conductivity	43.7 W/mK
Through plane conductivity	18.5 W/mK
Glass transition temperature	200 °C

IRD
IRD Fuel Cell Technology ranges among the leading experts in the fuel cell business, and has been working with R&D, design and manufacturing of Fuel Cells since 1982.

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