Efficient Use of Resources in Energy Converting Applications

Grant Agreement number:	**303024**
Project acronym:	**EURECA**
Project title:	**Efficient Use of Resources in Energy Converting Applications**
Funding Scheme:	**Collaborative Project**

1. Publishable Summary

1.1 WP2

The main objective of the Work Package 2 was to develop and produce new anode catalyst showing high tolerance to CO, new membrane with improved conductivity and mechanical properties, and finally MEAs designed for the operation of a PEMFC in the medium temperature (MT) range (90 to 120 °C) for a µ-CHP system supplied with hydrogen rich gas coming from the reforming of natural gas.

In parallel to these technological developments, extensive studies have been conducted thanks to dedicated and advanced experiments and modelling in order to understand the operation and durability of MT PEM.

FORTH calculated the relative permeabilities of GDLs reconstructed by SEM images and data received from EURECA partners (Figure 1). Complex two-phase flow models accounting for water transport were developed and applied on the same reconstructed GDL configurations. Simulation results included the effect of hydrophobicity on the phase and relative permeabilities of the GDL, and the overall performance of both LT PEM and MT PEM configurations. The results of these simulations have been implemented in a global model of performance used to design the stack in frame of WP4.

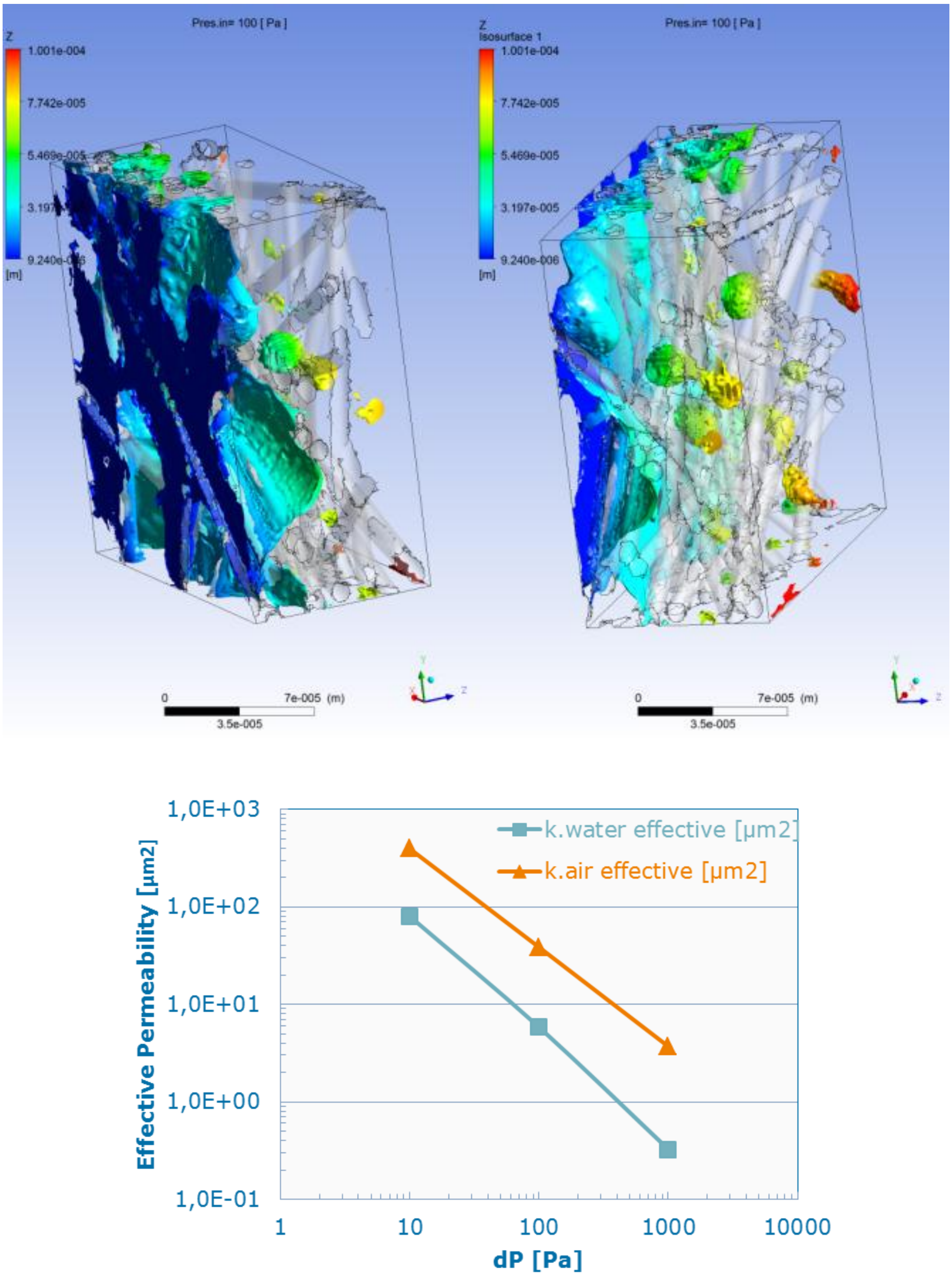


Figure 1. (Top) Complex simulations of two-phase flow in a reconstructed GDL. (Bottom) Effective, or phase, permeabilities of water and air at flow of water through hydrophobic GDL without phase change phenomena. Dependence of the phase permeabilities on the local pressure drop conditions for a volumetric saturation of 25 vol%.

An original proton conducting membrane made of a blend of a perfluorosulfonated ionomer (PFSI), such as Aquivion™, and a sulfonated hydrocarbon polyelectrolyte has been developed by CEA and show improved properties at 105°C compared to the bare commercial PFSI (Figure 2). We demonstrate that an optimal composition can be found in order to strengthen the mechanical properties of the PFSI while keeping its proton and water transport properties.

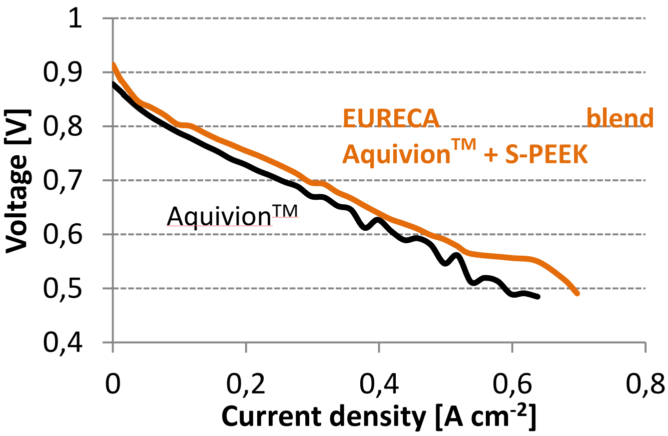


Figure 2. (Left) Picture of the original proton conducting membrane made of blend of Aquivion™ perfluorosulfonated ionomer (EW = 790 g mol^{-1}) and a sulfonated Poly(Ether Ether Ketone) (EW = 730 g mol^{-1}). (Right) Polarization curves obtained with the new blend in comparison with pure Aquivion™. 105 °C, H_2/Air 50 % RH, 1.76 bars st.1.2/2. Counter-flow.

University of Belgrade developed several types of catalyst supports with the aim of improvement in the HOR kinetics and CO tolerance of the anode catalyst, suitable for the operation of the MT-PEMFC stack in µ-CHP system**.** The novel PtRu/WOC anode catalyst exhibits higher mass activities towards the HOR in the presence of CO compared to the state-of the-art catalyst, which is due to the interactive nature of the catalyst support. This new anode catalyst allows enhancement of the CO tolerance to and the performance of the MEA above 90°C (Figure 3), compared to the state-of-the art anode catalyst. This catalyst has been produced and integrated in the MEAs of the final 83 cells stack which has been implemented in the final system.

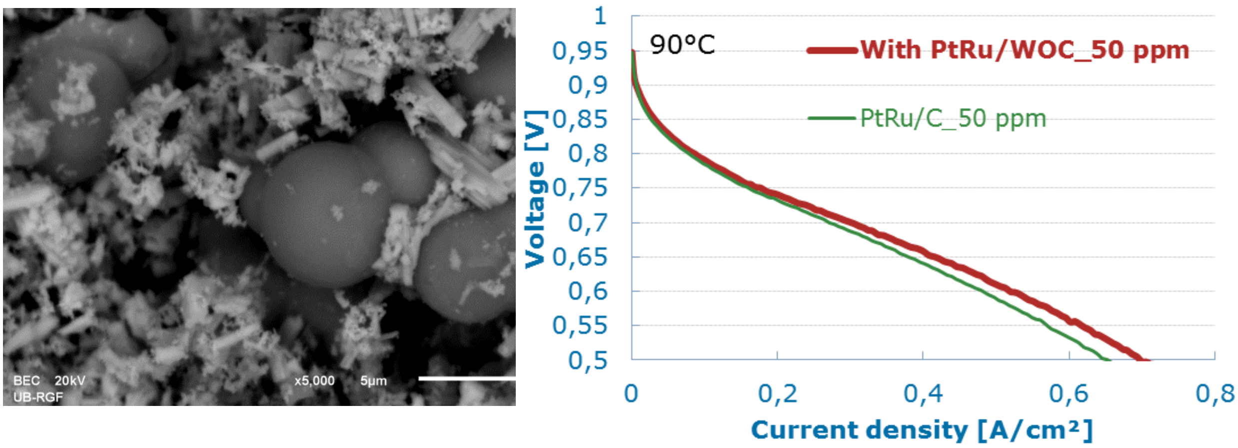


Figure 3. (Left) SEM picture of the new PtRU/WOC anode catalyst developed by University of Belgrade. (Right) Polarization curves obtained of MEA without (1st generation MEA – only commercial PtRu/C from Tanaka) and with PtRu/WOC (2nd generation MEA) anode catalyst. 90 °C_46/68 % RH, Reformate 50 ppm CO/Air, 1.2 bars, st.1.2/2. Counter-flow.

CEA developed three generation of MEAs within the frame of EURECA project with successive improvements. First generation integrated only commercial catalyst and membrane and was used for the initial steps of the developments of the MT stack. The new catalyst developed by University of Belgrade with enhanced tolerance to CO was implemented in the second generation MEA. This generation of MEA has been upscaled and produced by CEA in order to be used in the 83 cells full stack of the final system. The three generation of MEAs all exceed the target in term of performance at 90 °C (>0.3 W cm^{-2} at 0.5 A cm^{-2} or 0.6 V) and give better performance than all the state-of-the-art commercial MEA (Figure 4 and Figure 5). The 3rd generation MEA shows significantly enhanced performance, especially above 90 °C and it is the best MEA tested in all the operating conditions: >0.45 W cm^{-2} at 90 °C_46/68 % RH and >0.3 W cm^{-2} at 105 °C_40/26 % RH, at 0.6 V and 1.2 bars with 10 ppm CO in the reformate. This type of MEA has been integrated in a five cells stack which has been tested in the medium temperature range, up to 105 °C.

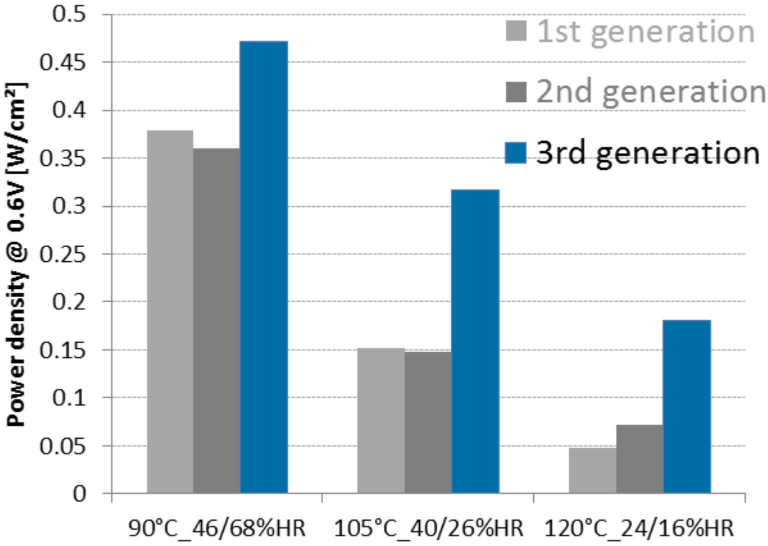


Figure 4. Power density in different operating conditions with the 1st, 2nd and 3rd generation MEA developed within EURECA. 90 °C_46/68 % RH, 105°C 40/26 % RH, 120 °C 24/16 % RH. Reformate 10 ppm CO/Air, 1.2 bars, st.1.2/2. Counter-flow.

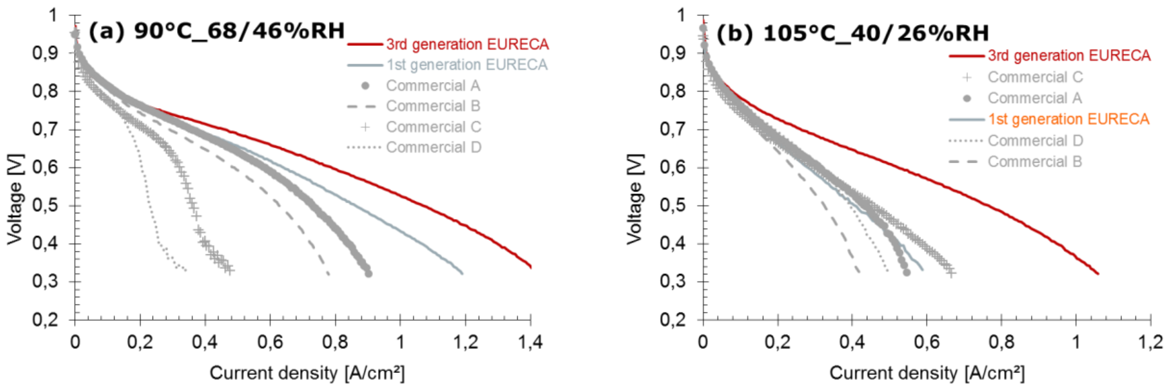


Figure 5. Polarization curves in different operating conditions with commercial MEAs and with 1st and 3rd generation EURECA MEAs. (a) 90 °C_46/68 % RH, (b) 105 °C 40/26 % RH. Reformate 10 ppm CO/Air, 1.2 bars, st.1.2/2. Counter-flow.

The durability of the MEA developed in EURECA is less than 3 % over 1500 hours at 90 °C, which corresponds to a decay around 15 µV h which is a value similar to those reported for low temperature PEMFC (<80 °C) (Figure 6).

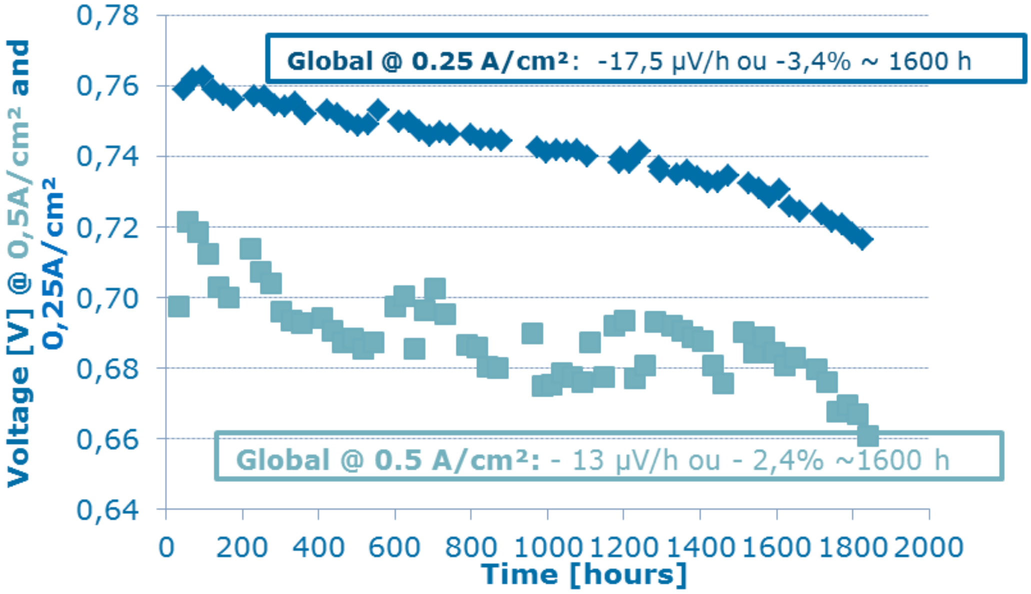


Figure 6. Evolution with time of the voltage recorded with 1st generation MEA during durability test with the load profile (0.25 and 0.5 A cm^{-2}) of the µ-CHP application, 90 °C Reformate 10 ppm CO/Air, st.1.2/2. Counter-flow.

The durability tests surprisingly show that the decay in performance is much more pronounced at 90 °C than at 105 °C with 50 ppm CO in the reformate. This has been ascribed to the enhanced tolerance to CO poisoning as temperature increases, showing the advantage of medium temperature operation when using high level of pollutants at the anode. Advanced TEM analyses have been conducted by CEA in order to characterize the evolution of the microstructure of the MEA during durability tests, and to understand the causes of the decrease in performance. They reveal that commercial PtRu/C catalyst is unstable. Ru is dissolved and precipitates within the membrane and also in the microporous layer at the anode side, depending on the local operating conditions, especially on the presence of liquid water (Figure 7). The degradation of the anode catalyst can explain the degradation rate observed in presence of CO.

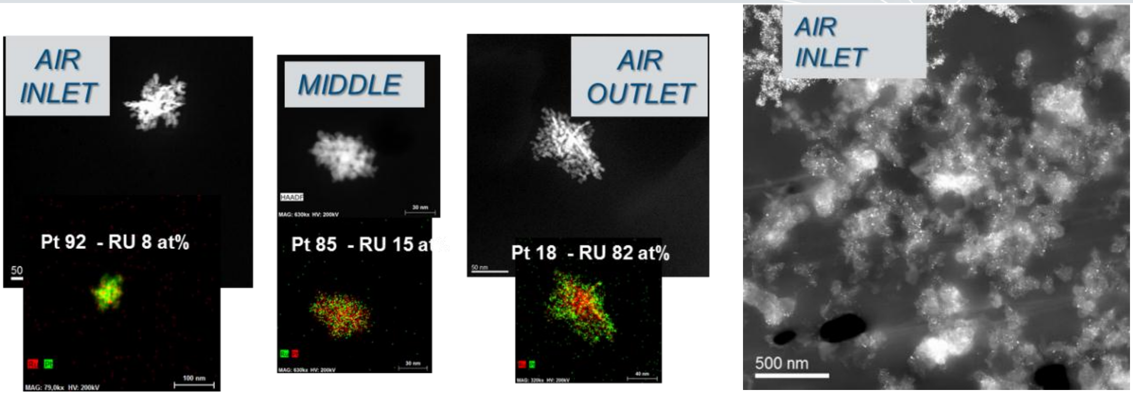


Figure 7. (Left) Images and chemical compositions of precipitates within the membrane in different areas of the MEA, with respect to the gas inlets/outlets, (Right) Images of precipitates (white dots) of Ru within anode microporous layer (MPL). 90 °C_46/68 % RH, Reformate 10 ppm CO/Air, 1.2 bars, st. 1.2/2. Counter-flow.

A lifetime estimation model has been developed by University of Belgrade in order to predict the durability of the MT stack. It was shown that this numerical model can be used to accurately predict the FC performance over time (Figure 8), especially when the dynamic load of the stack is planned, which is consistent with the operation of the µ-CHP system in the household.

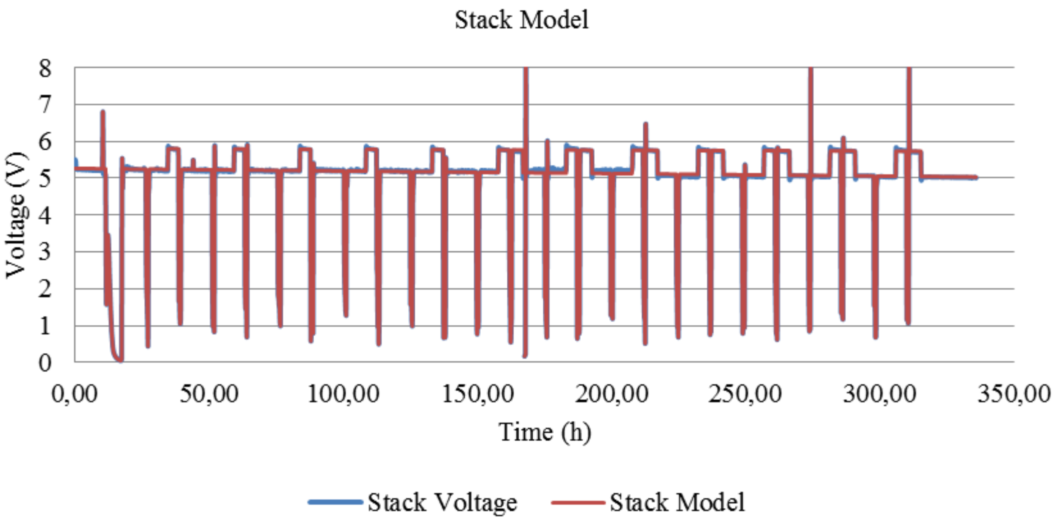


Figure 8. Evolution of experimental and simulated of stack voltage during durability test.

1.2 WP3

Bipolar plate development:

In the project EURECA graphite composite based bipolar plates and gaskets for MT-PEM (up to 120 °C) fuel cells have been developed, manufactured and implemented in the EURECA stack and fuel cell system. Regarding the bipolar plate, state of the art where the project started was a consistent manufacturing process for LT-PEM materials with polypropylene binder and a successful proof of concept for MT-PEM bipolar plates based in the fluoro-polymer PVDF (polyvinylidene-fluoride) as a binder. Within the first half of the project, the partners developed PVDF bonded bipolar plates and characterized them with respect to electrical conductivity and mechanical stability at the anticipated temperatures. The heat deflection temperature of pure PVDF polymer is in the area of 115 to 148 °C, depending on the test method. Due to graphite fillers the heat deflection temperature usually increases by approximately 20 °C, thus the theoretical values propose that PVDF may fulfil the thermomechanical requirements of MT PEM fuel cells. This was confirmed by the experiments in EURECA project. Therefore, the partners developed a tailored thermomechanical teste, where a mechanical load corresponding to the real conditions of a fuel cell stack is applied on a blank plate (here: 10 kg steel block on 1 cm^2 plate sample, equivalent to 100 N cm^{-2} pressure). The temperature was 120 °C with a duration of approximately 100 hrs. All dimensional changes and weight changes have been measured. As shown in Figure 9, several PVDF-based plates remained constant in the grey area of +/-1 % in height, width or thickness, which is the experimental resolution. Thus, these PVDF based plates passed the test and showed sufficient mechanical stability under the anticipated operating conditions.

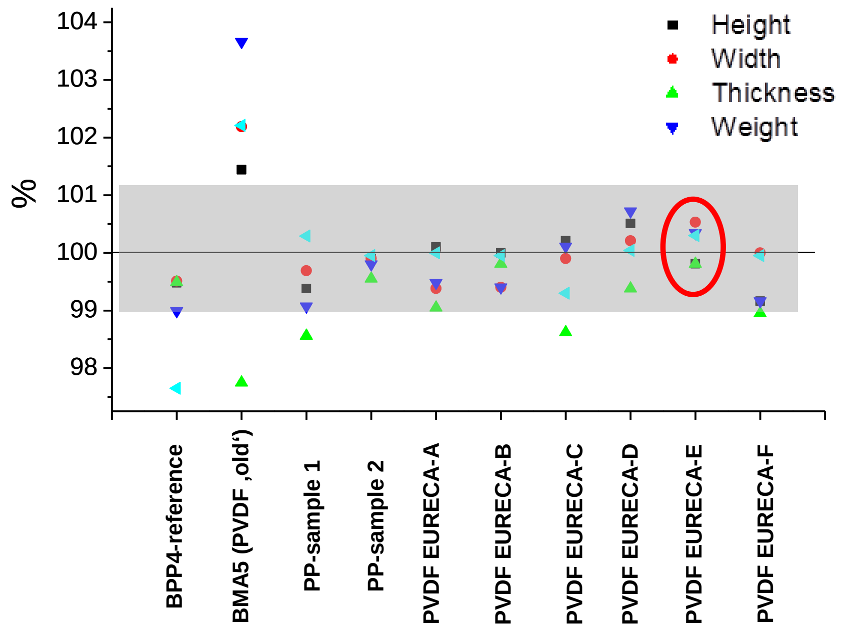


Figure 9. Thermo-mechanical test results of different bipolar plate materials. BPP4=high temperature PEM material; MA5 = old PVDF configuration before EURECA; PP=polypropylene bonded LTPEM plates; PVDF Eureca = materials developed in this project. All samples with dimensions remaining in the grey area are in-spec

A broad variety of PVDF bonded compounds has been synthesized, and the corresponding plates have been manufactured and tested. The aim was to achieve plates with a sufficient electric conductivity. Obviously, there is a strong influence of compounding and moulding conditions on the physical properties of a plate. The partners learned that bipolar plates with very similar or even identical chemical composition may strongly differ in der function. An impression of this work is given in Figure 10, where a selection of plate configurations is presented. Three configurations passed the milestone MS 3.1. of <10 µOhm m electrical resistivity, however only one them had been selected for the project (sample 'E'). The partners decided for the type 'Melange-E' because if the reproducibility of conductivity and the fact, that 'E' does not contain functional carbon additives, which may complicate the compounding process.

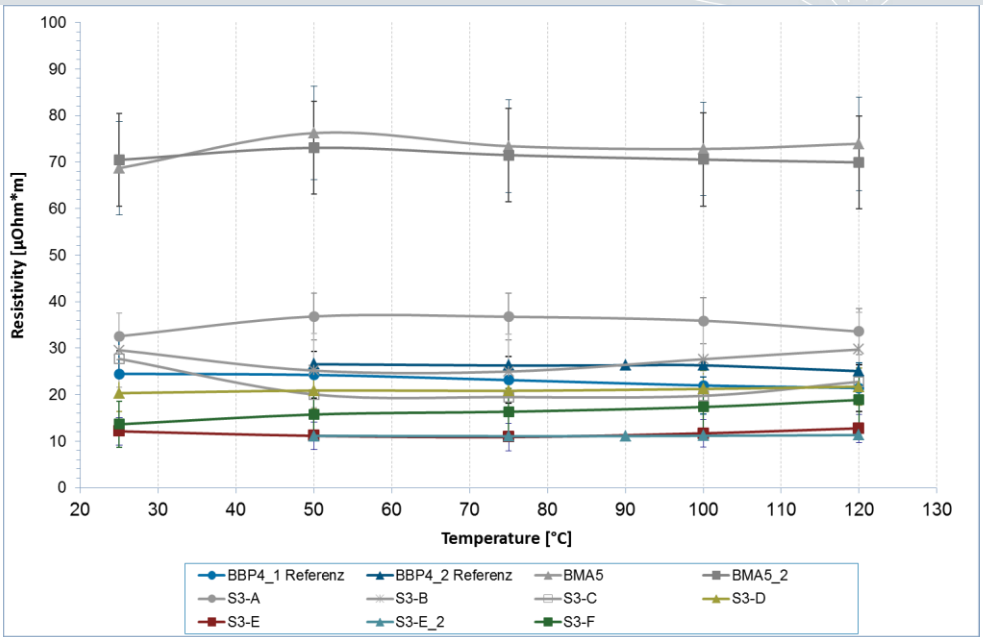


Figure 10. Electrical resistivity as a function of temperature for a variety of PVDF based plate materials. Blue: reference BPP4 commercial material from Eisenhuth; grey: new materials with low conductivity; colourful: successful samples, here S3-D, E and F with low resistivity, < 10 µOhm*m. The best material fulfils MS3.1 within the experimental resolution. The sample S3E (type 'Melange-E') has been selected for EURECA plate manufacturing (test data by Fraunhofer ISE).

Starting point of EURECA's plate development based on PVDF binder was the configuration 'BMA5', which is mentioned in Figure 10 with its too high resistivity of 70 µOhm m. There was evidence that conductivity can be improved in PVDF based plates due to a pre-screening, however this was not reproducible and not at all a stable and mature manufacturing process. Within EURECA several parameters, such as composition, PVDF-types (powder or pre-grinded pellets), conductive carbon additives, carbon types and process parameters of compounding (adding components, stirring, extrusion) have been varied to figure out highly conductive and reproducible configurations. After making the decision for the type Melange-E, the partners focused on reproducibility, process consistency and quality control testing of this material. Several runs on a technical scale have been performed and each sample has been characterized. Particular attention has been paid on material homogeneity, which is characterized by a conductivity mapping of each plate. Therefore, the surface conductivity is determined at 26 data points per blank plate. A criterion of success is not only the conductivity itself, but also the range covered by the test values. The smaller the range is the more homogeneous the material. The process control of melange E could be significantly improved so that for example within 52 plates only one or two plates are out of spec, as shown in Figure 11.

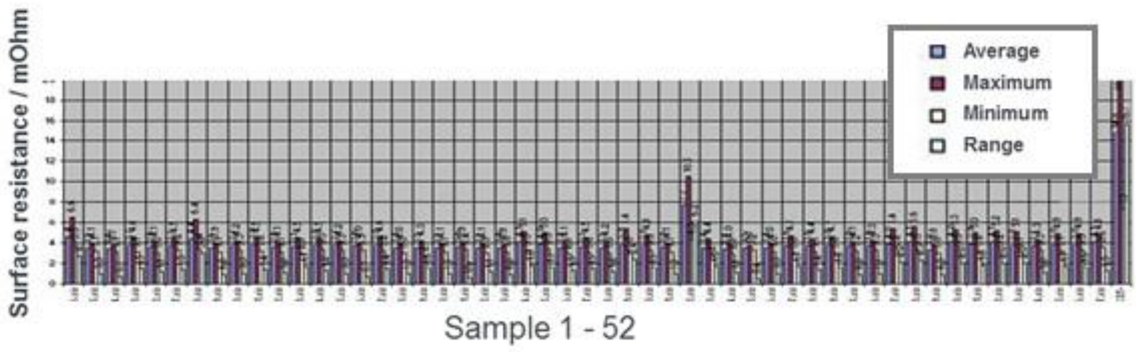


Figure 11. Conductivity mapping of PVDF (type Melange-E) blank plates, 52 plates in test series #2. Results show a good consistency with 50 plates in spec (homogeneous) and two plates out of spec. All surface resistance values are relative numbers (only comparable within one plot)

Based on this process (both in the final stage and in earlier stages) the bipolar plates for testing purposes, several short stacks and the full size EURECA stack have been manufactured and delivered to the relevant partners. Finally, with 'Melange-E', a new PVDF based bipolar plate configuration has been set up, both chemically and mechanically resistant to up to 120 °C, fulfilling the electric conductivity requirements of the project and being manufactured on a technical scale with reproducible and consistent quality. Careful analysis and large experimental efforts have been spent by several partners in QC testing of plates with respect to their dimensional tolerances. Critical points were the sealing groove depth and the overall plate thickness. The relevant partners agreed to defined procedures and points on the plates where the parameters have to be measured. Finally good progress was made to synchronize the testing in different labs.

In parallel, at the interface to WP4 'stack development', simulations of the flow field in the interior of the gas channels have been performed. Using the input of stack and MEA manufacturer, a computational model has been developed which allows a mapping of the pressure distribution along the gas channels. The corresponding flow field has been calculated based on experimental data from operating conditions, such as pressure, stoichiometry, and intended design of the bipolar plates. In general, this modelling enables to visualize potentially blocked channel structures, which may cause increased pressure gradients and decreased velocities. The flow field, which is implemented in the stack, was found to provide an even distribution of gases without major flow inhomogeneity. Results have shown that the effect of geometry is not linear with the velocity field, and that scaling of the geometries at the depth dimension (through-plane) can be considered as promising result for the optimization of the flow structures by the stack manufacturer. Additionally, the theoretical work combined with experimental stack testing of WP4 strongly contributed to a fundamental understanding within the consortium regarding the processes in MEA and flow field and the functions of each component under MT-PEM conditions. This contributed directly to generating optimised geometries with improved gas flow and mixing conditions.

Gasket development:

WP3 covers also gasket development for the MT-PEM stack. Assuming an upper temperature limit of 120 °C for long term applications, in the first half of the project the partners focused on fluoro-elastomers of the FKM family. Due to the redefinition of temperature targets towards 90 °C, also temperature resistant silicon based gaskets became an alternative. Within the project, both routes have been successfully finalized. The material qualification for MT-PEM fluoro-elastomers (FKM types) has been concluded. Finally, with the anticipated operating temperature of 90 °C the partners selected a silicon type gasket. Despite the FKM gasket is definitely superior with respect to chemical and thermal stability, the less stable, but more cost-effective silicon is softer and compensates dimensional tolerances of MEAs and bipolar plates. The gaskets for the full size EURECA stack have been manufactured and implemented in the stack. Additionally the partners developed a proof of concept for integration of gaskets directly on the MEA. A silicon based gasket has been applied on the GDL / membrane edge of the MEA, providing a ‘7-layer-structure’. The MEA/Gasket assembly has been ex-situ tested for gas tightness in a test cell, were the pressure can be reduced on one side and the pressure as a function of time can be detected. Within the experimental resolution the MEA with integrated gasket was gas tight. This work has been performed in parallel to MEA development with the target to demonstrate a proof of concept only. Due to strong interactions of GDL and membrane properties with the gasket integration it would have complicated the MEA development unnecessarily if integrated in the EURECA full size stack.

1.3 WP 4

The goal of WP4 was the development of a MT-PEM fuel cell stack based on the cell and stack design from the INHOUSE LT-PEM fuel cell stack. This development was guided by modelling data of flow rate and concentration distribution of the reactants and fluids made by FORTH and in addition by investigations using a segmented single cell made by FRAUNHOFER ISE. This new developed stack design was approved in performance and degradation tests of several short stacks with cell quantity of 5 to 8 cells (active area ~200 cm^2 per cell, Figure 14, right) equipped with 3 generations of MEA developed in WP2. At the end a full scale stack with 83 cells was realized (Figure 12, right) and used for the MT-PEM system integration done in WP5.

At the beginning of EURECA project specification of MT-PEM process parameters were defined in WP4 based on

- DoW targets
- Options of MEA development in WP2
- Literature study
- System prospects and limitations especially anodic gas composition, gas pressures and air humidification

The targeted process parameters have been strictly used for

- Development of MT-PEM-MEA in WP2
- Single cell testing in WP2
- Development of MT-PEM-Stack in WP4
- Stack testing in WP4
- Development of system in WP5
- System testing in WP5

The “transfer” of the commercial LT-PEM Stack design to a MT-PEM Stack required different development work shown in Figure 12.

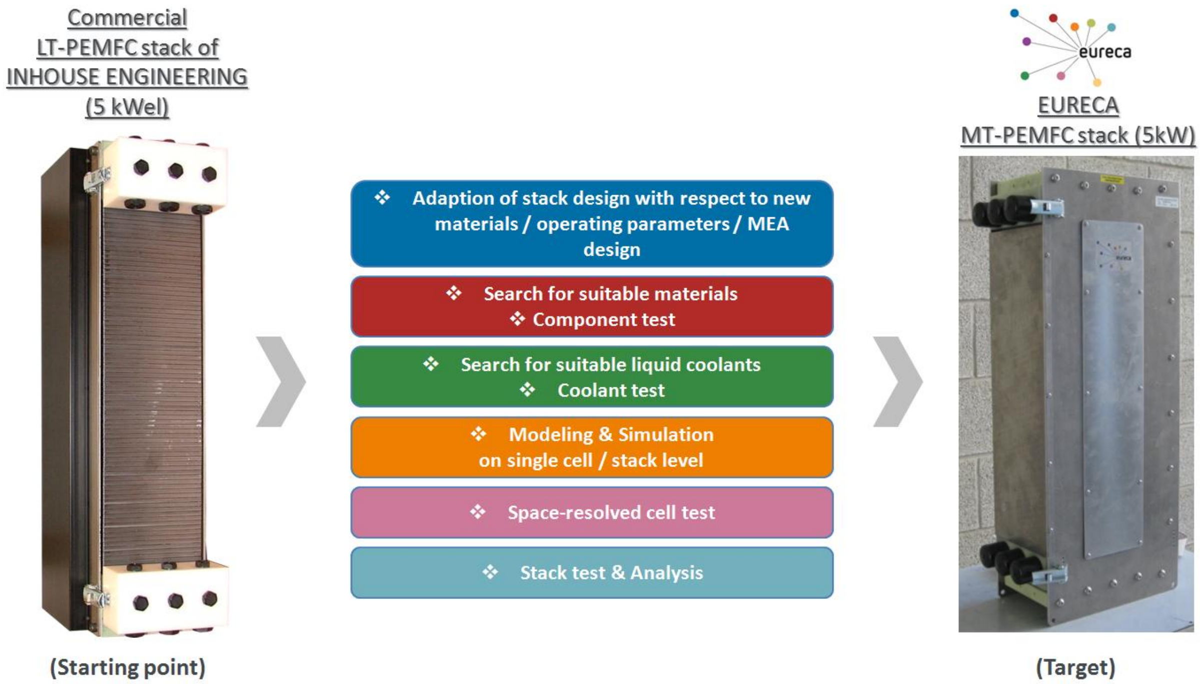
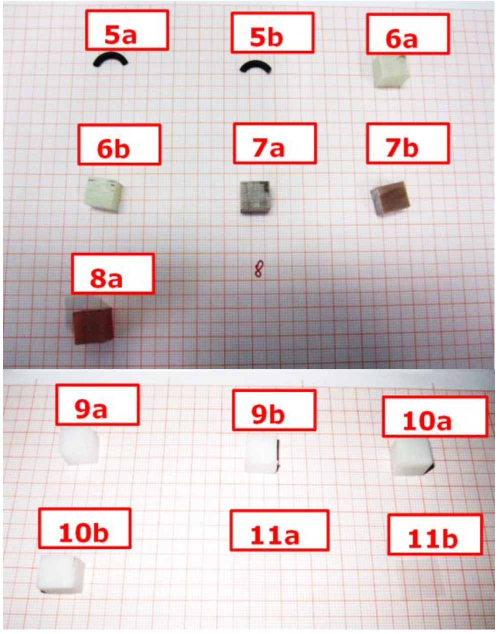
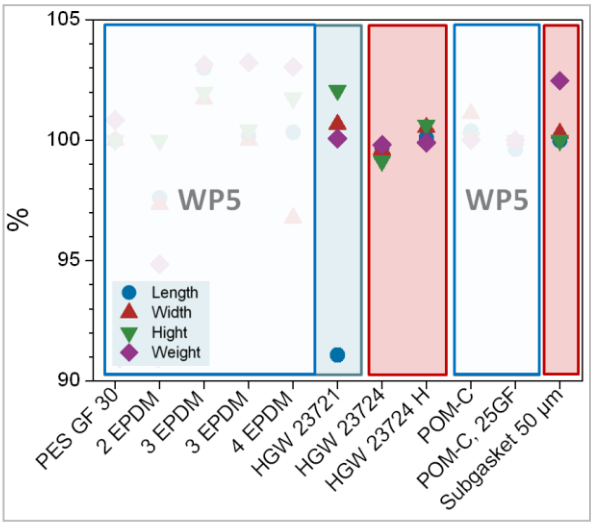


Figure 12. Development work for transfer of LT-PEM stack into MT-PEM stack

1.3.1 Adaption of stack design

The general adaption of the stack design was characterized by MEA integration into available LT-PEM stack design as well as the usage of suitable materials like for gaskets, graphite plates and end plates with respect to the defined MT-PEM process parameters. Due to the increased temperature level (90 – 120 °C) also a suitable liquid coolant had to be found which influenced the stack design. Regarding the new stack materials extensive material tests were conducted on MT-PEM operating temperature level in contact with new synthetic cooling media which provided useful results for selection of stack materials as well as BoP component materials of the system (Figure 13).





Blue – 95 °C, 200h
Red – 120 °C, 200 h

Figure 13. Selected material samples of material test with synthetic cooling medium (Material test at NEXT ENERGY)

For the determination of the optimal and well distributed compression of the MEA (for best performance) and the gasket as well (for leakage free stack) several preliminary investigations were done like determination of real thickness and thickness distribution of the project MEA from different batches and different producers (CEA / CEGASA), CAD analysis of optimal/recommended compression rate of MEA and gaskets, compression tests with samples of project MEA (Figure 14, left), compression tests with gaskets and also leakage tests with single cell units. After completion of preliminary tests several MT-PEM stacks (6 short stacks & 1 full stack) have been assembled in EURECA project, all with the determined optimal clamping force. Figure 14, right shows one of the MT-PEM short stacks.

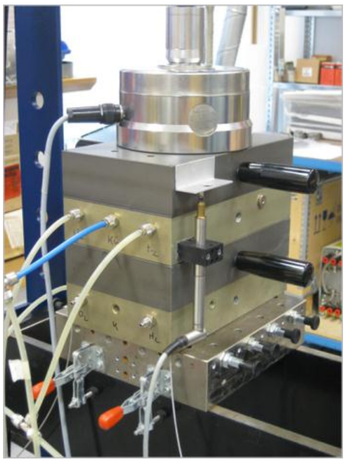


Figure 14. Compression test set-up for single cell compression with force and way sensor (left), MT-PEM Short stack with 8 cells (right)

In the frame of usage of a new synthetic coolant it was necessary to adapt the cooling channels inside the liquid cooled stack. Several studies and experiments were done to compare the behaviour of the new coolant with water. It was found out that the pressure losses between the inlet and the outlet of a stack are higher for the synthetic coolant (~400 mbar) than for water (~275 mbar). Furthermore it was found out that a higher coolant flow rate is needed for same cooling conditions as for water which led to a much higher pressure loss (~750mbar). By increasing the cross section of the cooling channels the pressure loss of synthetic coolant (~250 mbar) could be reduced to a common level like for using of water (Figure 15).

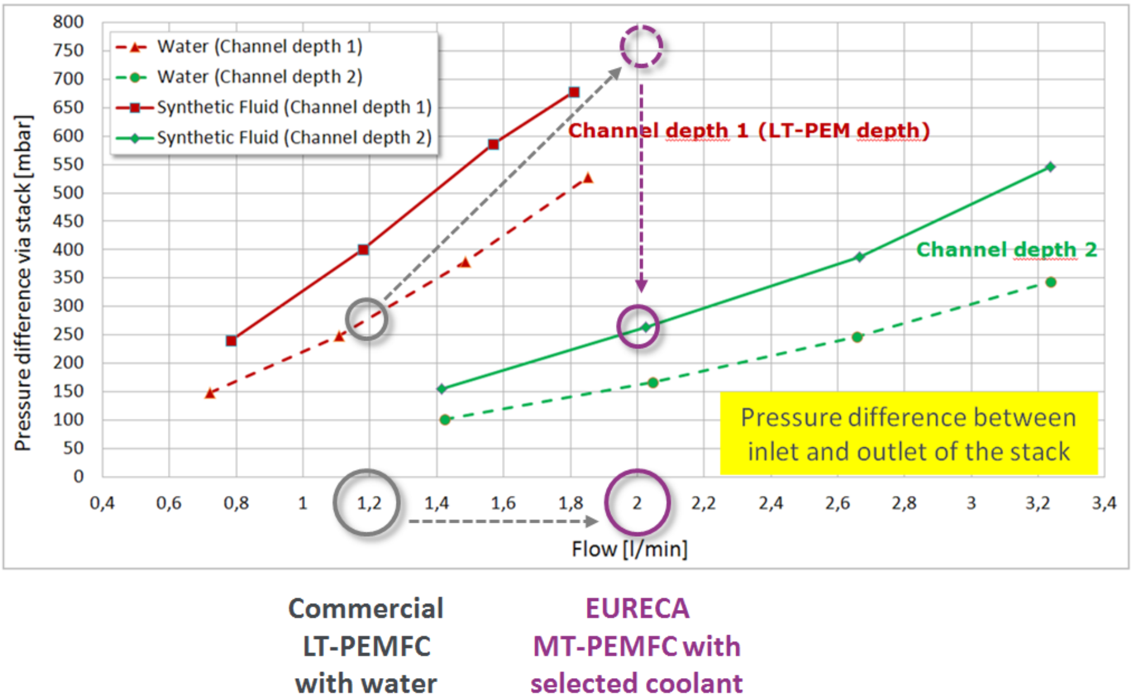


Figure 15. Adjustment of cooling channels inside the stack

Beside the optimal MEA integration into existing LT-PEM stack design and the other mentioned adaptions the overall stack development shown in Figure 16 was structured in:

- Short stack tests with different generations of MEA
- Optimization of gas channel structures
- Assembling of full stack for the system

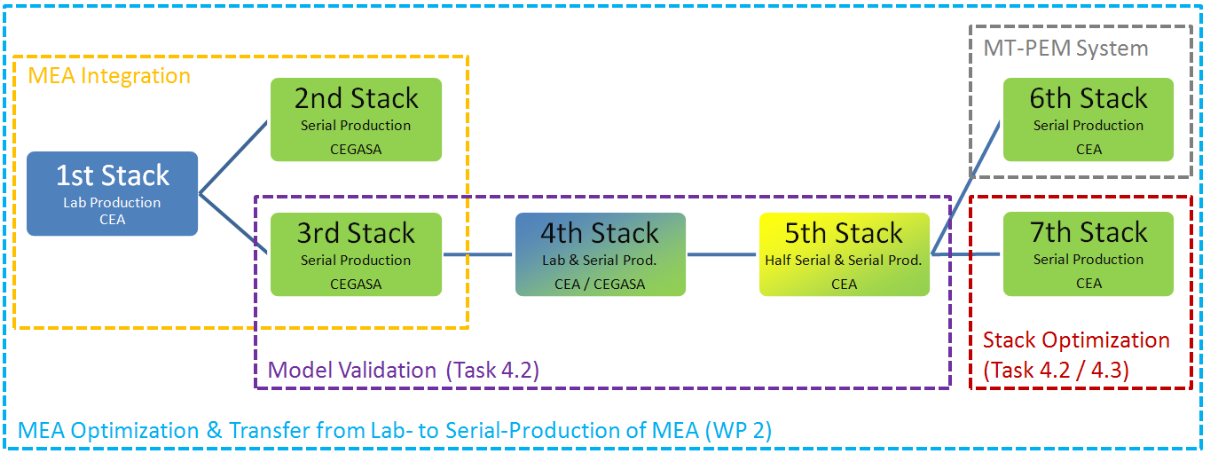


Figure 16. History of stack development

1.3.2 Stack test & Analysis

In the frame of WP4 all stacks developed and assembled have been tested at FRAUNHOFER ISE or NEXT ENERGY and INHOUSE (function test / break-in). The approach was to share the stack tests to spare time, to use different possibilities of the institutes and also to perform same tests and to compare the results. Based on FCTESTQA new performance, stability and durability test routines and common definitions were developed by INHOUSE, NEXT and FRAUNHOFER ISE to optimize operating parameter. This routines and the optimization process were successfully validated by NEXT ENERGY, ISE and INHOUSE with the LT-PEM stack of INHOUSE at first. In the following the basic tests are listed:

INHOUSE:

- Function test (70 °C)
- U/I characteristic at Begin-of-Life (70 °C)

FRAUNHOFER ISE:

- U/I characteristics at Begin-of-Life / Begin-of-Test (70 °C/90 °C)
- U/I characteristics for different variation protocols (humidity, stoichiometry, pressure, stack temperature)
- Stability tests (≥100 h at constant load, several periods)
- EIS spectra / HFR of stack
- EIS spectra / HFR of each cell

NEXT ENERGY:

- U/I characteristics at Begin-of-Life / Begin-of-Test (70 °C/90 °C)
- U/I characteristics for different variation protocols (humidity, stoichiometry, pressure, stack temperature)

- Stability tests (≥100 h at constant load, several periods)
- Special durability test with start/stop integration
- CO tests

In the frame of stack testing institutes FRAUNHOFER ISE and NEXT ENERGY have upgraded their test stands for using of synthetic cooling medium based on perfluorinated polyether up to 120 °C as well as for supply of the stacks with defined synthetic reformate. Furthermore NEXT ENERGY has implemented a possibility to realize CO concentrations up to 50ppm.

Figure 17 shows the inner part of the institutes' test stands with integrated EURECA short stacks.

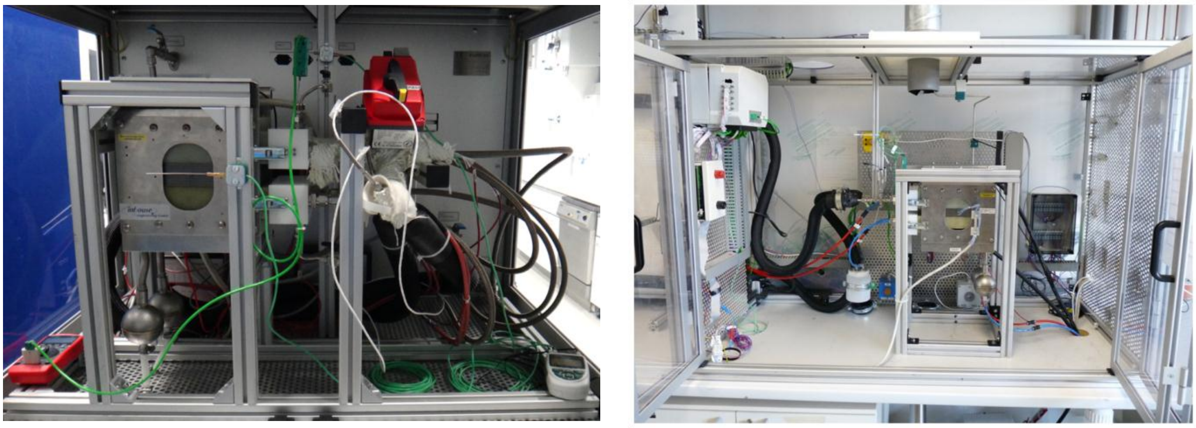


Figure 17. Inner part of the test bench at NEXT ENERGY (left) and at FRAUNHOFER ISE (right) with integrated EURECA stacks

Figure 18 shows exemplary for all tested short stacks the complete stack operation history of the final EURECA 5-cell-short-stack with 3rd generation MEAs at FRAUNHOFER ISE including the execution of U/I-characteristrics depending on stoichiometries, dew points and gas pressure as well based on protocols defined in the project. Several stability test periods were carried out at constant load of 80 A (0.41 A cm^{-2}) with Begin-of-Test and End-of-Test polarization curves in-between.

The cell performance at Begin-of-Life reached an average cell voltage of $U_{Av(1-5)}$=660 mV @ 0.51 A cm^{-2} with very low deviation between the cells (25 mV). The cell voltage to reach a power density of 300 mW cm^{-2} is around 680 mV. The final short stack meets the targets of EURECA very well.

A degradation rate of approximately 0.094 mV per hour was calculated by FRAUNHOFER ISE after approximately 535 hours of run time.

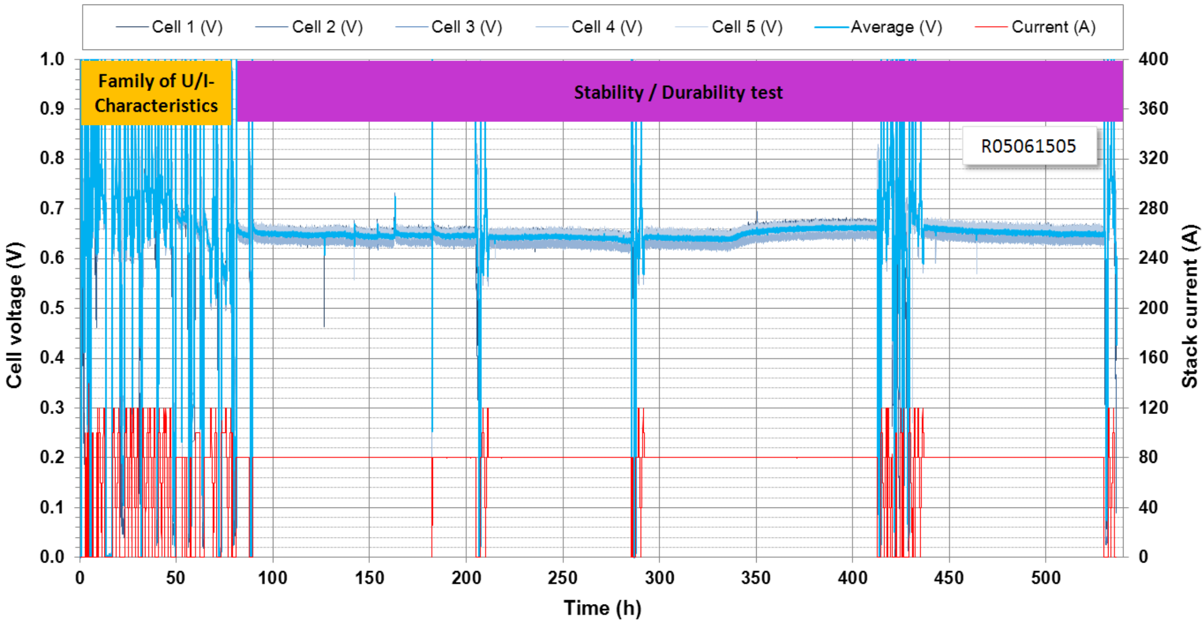


Figure 18. Stack operation history (time of real operation) of 7th EURECA stack at MT-PEM conditions (~90 °C) at FRAUNHOFER ISE

The results of the stack temperature variation, done with steps of 70 °C, 90 °C and 105 °C stack temperature (coolant outlet), are given as U/I/P plots (Begin-of-Life) in Figure 19. The best polarization curve was determined at 70 °C with a performance loss of ~9 % at 100 A (0.51 A cm^{-2}) with stack temperature change from 70 °C to 90 °C and a further loss of ~14 % at 100 A (0.51 A cm^{-2}) with change from 90 °C to 105 °C.

The calculated power densities are also given in the figure. Exemplary highlighted is the power density of 300 mW cm^{-2} which was targeted in EURECA. This level was reached at stack temperature of 90 °C at an average cell voltage of 0.680 V and at stack temperature of 105 °C at an average cell voltage of 0.540 V.

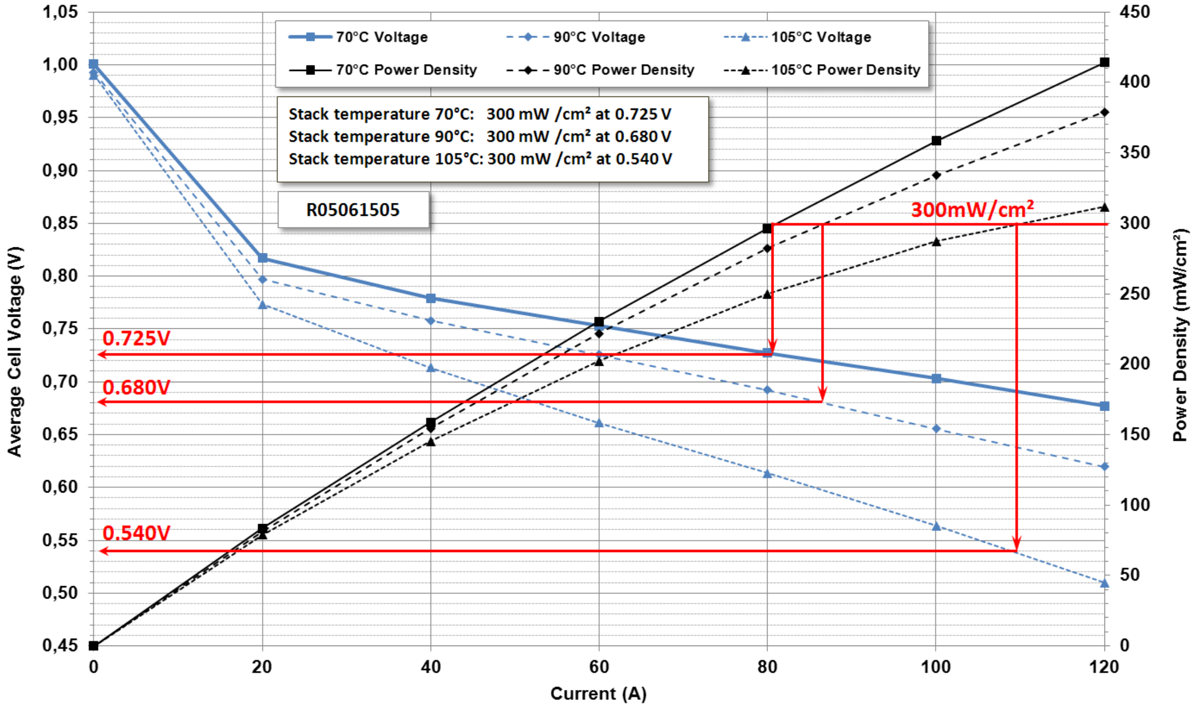


Figure 19. Temperature variation of 7th stack (3rd generation MEA) at Begin-of-Life at FRAUNHOFER ISE
Gas composition An/Ca: 75 % H_2 + 25 % CO_2 + 0 ppm CO
Temperature gas inlet An/Ca: 61/61 °C | 81/71 °C | 81/71 °C
Dew point An/Ca: 60/60 °C | 80/70 °C | 80/70 °C
Temperature coolant inlet/outlet: 65/70 °C | 85/90 °C | 100/105 °C
Coolant: Galden® HT 170

After regular test at FRAUNHOFER ISE the stack test was continued at NEXT ENERGY. The Figure 20 shows the CO test protocol at constant load of 80 A (0.41 A cm^{-2}). A variation of CO concentration 0 ppmv, 10 ppmv and 50 ppmv was done. On each CO concentration step the stack was operated for ~24 hours. Between 0 ppmv and 10 ppmv there was an automatic test stand shut-down caused by a dysfunction.

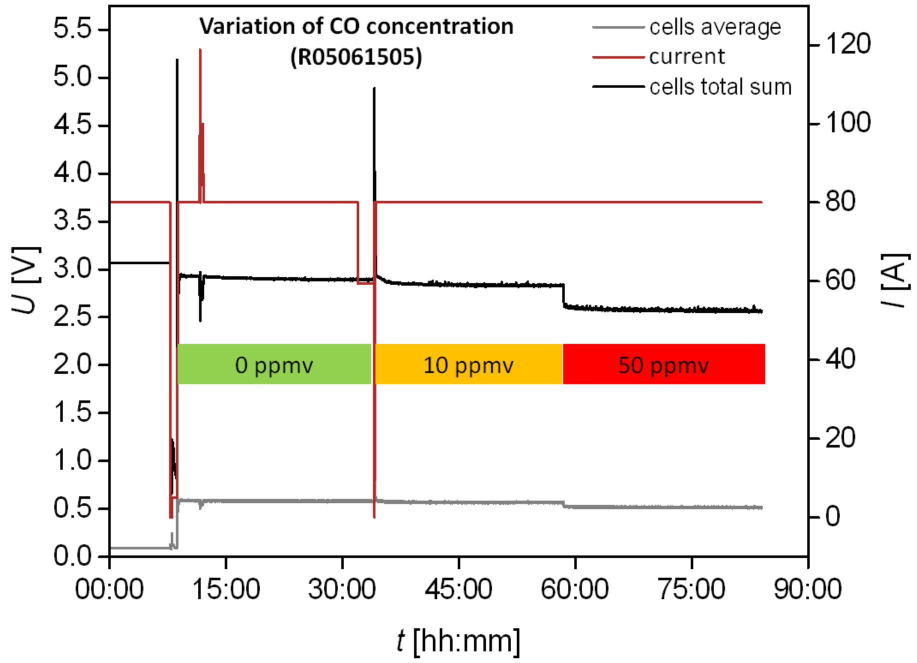


Figure 20. CO test of 7th EUREKA stack (3rd generation MEAs) at NEXT ENERGY
Process values:
Stack current: 80 A (0.41 A cm^{-2})
Gas composition An/Ca: 75 % H_2 + 25 % CO_2 + 0|10|50 ppmv CO / Air
Stoichiometry An/Ca: 1.3/2.0
Temperature coolant outlet: 90 °C
Dew point An/Ca: 80 °C/70 °C
Coolant: Galden® HT 170

Following figure shows the influence of CO concentration on the stack at 80 A (0.41 A cm^{-2}). The values are mean values of all cells and the last 8 hours of each concentration step. The average cell voltage started with 580 mV at 0 ppmv CO and decreased to 568 mV at 10 ppmv CO and 517 mV at 50 ppmv CO. Expressed as percentage decrease the voltage loss is ~2 % at 10 ppmv and ~11 % at 50 ppmv.

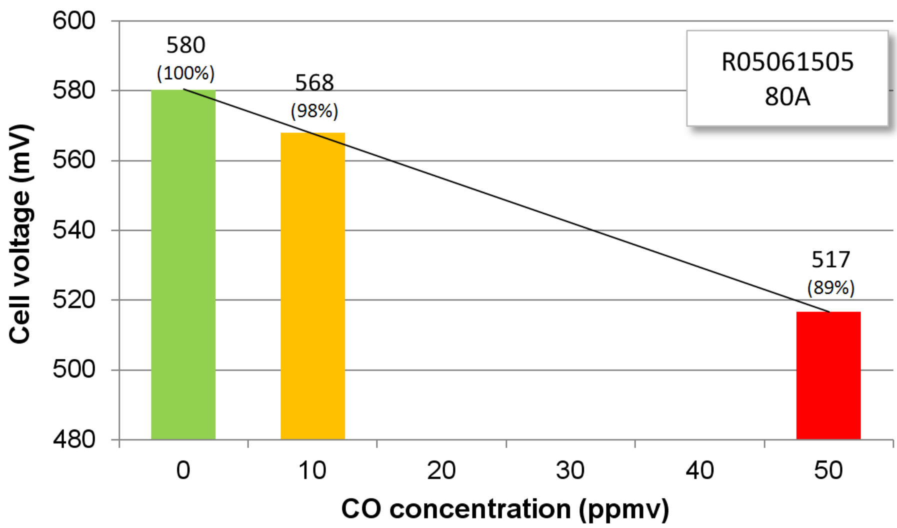


Figure 21. Results of CO test of 7th EURECA stack

Furthermore it seems that there is a linear trend of voltage decrease in dependence of CO concentration with approximately 12.8 mV loss per 10 ppmv CO.

Figure 22 compares Begin-of-Life (BoL) polarization curves of different EURECA stacks (Stacks 3 – 7) equipped with the different generations of MT-PEM-MEA. All stacks were operated with synthetic reformate (75 % H_2+25 % CO_2) but the 6th stack was operated with real reformate. The progress of MEA development is in evidence. The 1st generation MEA (3rd and 4th EURECA stack) gave the lowest cell voltage with strong deviation of the cells (visible due to error bar). 2nd generation MEA with PtRu on WOC (from University of Belgrade) for the anodic electrode and used for the 5th and the 6th EURECA stack showed slightly better results especially regarding the cell voltage deviation. The curve for the 6th stack is based on an 83 cell stack for the system. The shown polarization curve of 6th stack is lower than the curve of 5th stack but it has to be taken into account that the curve of 6th stack was performed in the EURECA system with real steam reformate based on natural gas type H. The final EURECA stack number 7 includes all final EURECA development status of MEA (WP2, 3rd generation), graphite material (WP3) and optimized channel structures (WP3, WP4). The U/I characteristic of 7th stack with 3rd generation MEA was the best of all EURECA stacks.

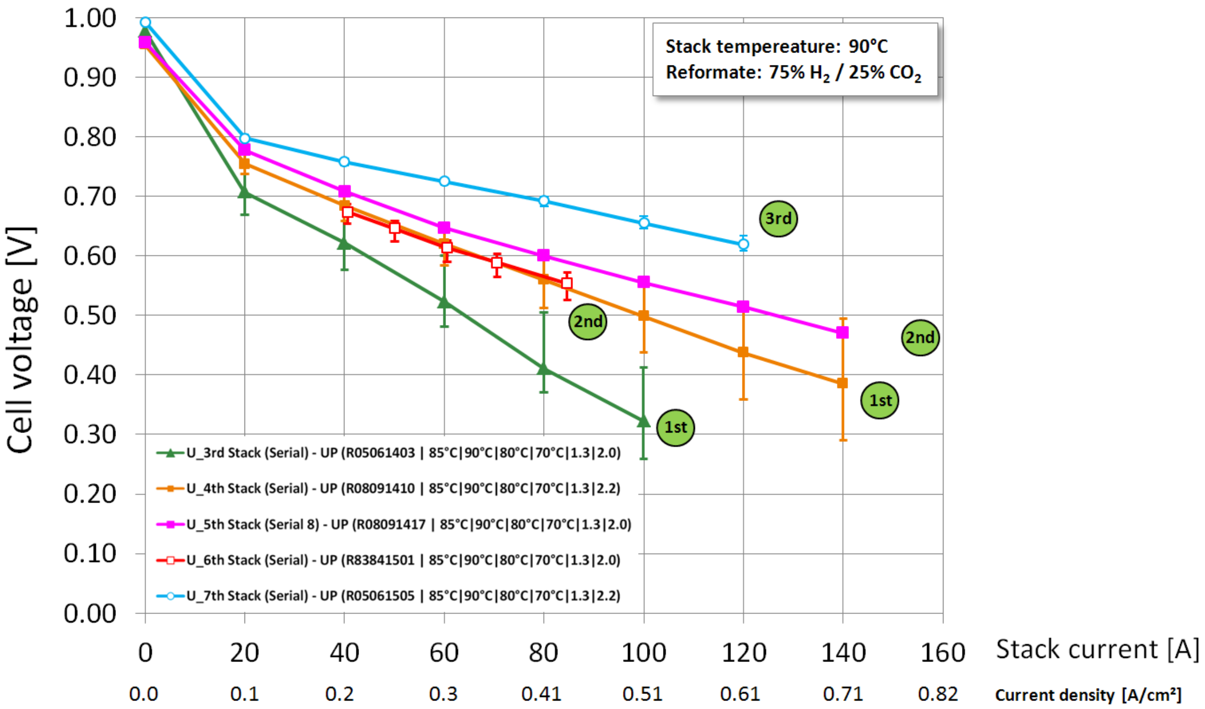


Figure 22. BoL Polarization curves of EURECA stacks with 1st, 2nd and 3rd generation MEA (from serial production process)

1.3.3 Segmented cell

FRAUNHOFER successfully implemented its unique technology of segmented impedance measurement into the stack design of INHOUSE for MT-PEM application. Figure 23 shows the segmentation of the cell and the contacting pins (left) as well as the Multichannel characterization system devices (right).

Figure 23. Segmentation and contacting (left), Multichannel characterization system devices (right)

The cell was divided into 64 segments with different area sizes. The target was to get knowledge about space-resolved results of EIS and HFR segment by segment and not least to localize problems over the active area.

Following Figure 24 shows exemplary results of segmented cell by using the 2nd generation MEA type. On the left side the EIS of all segments are shown for the process values and MEA type given beside the figure. On the right side of Figure 24 the voltage distribution is shown. It can be seen that the voltage was higher near the inlet compared to the outlet and there is a clear gradient. This is due to the fact that the performance of the cell is dominated by the concentration of cathode gas (oxygen from air) which gets depleted from inlet to outlet.

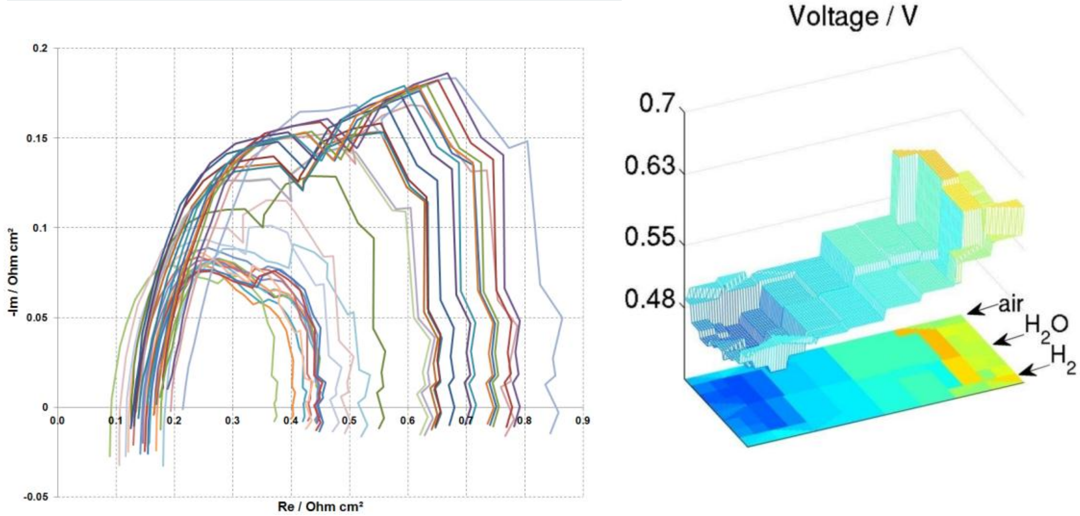


Figure 24. **Electrochemical impedance spectra (left), Voltage distribution (right)**
Process values:
Galvanostatic operation (0.51 A cm^{-2} for each segment)
Gas composition An/Ca: 75 % H_2 + 25 % N_2 / Air
Stoichiometry An/Ca: 1.3/2.2
Temperature coolant outlet: 86 °C
Dew point An/Ca: 74 °C
MEA type: 2nd generation, Serial produced by CEA (same MEA as used for the full 6th stack of EURECA system)

The segmented cell gave important results for the development of the optimized channel structures which are similar to the LT-PEM channel structures but with some modifications which eliminate local problems of stack internal gas transport.

From segmented cell test data it was concluded that the chemical reaction at the inlet seems to be good (highest voltage, small HFR, small 1st loop of EIS, small 2nd loop of EIS). Near the outlet, mass transfer limitations occur (smallest voltage, small HFR, small 1st loop of EIS, larger 2nd loop of EIS).

These results have been used for the optimization of gas channel structures. The gas channel cross section of air channels was reduced to increase the gas flow velocity of the cathode. The cross section of the anode channels in the outlet region was increased to avoid droplets of liquid water inside the anodic channels.

1.3.4 Modelling and simulation

A single Fuel Cell model was developed that takes into account transport of species across the different parts of the unit, electrochemistry in the catalyst layer and temperature variation. Realistic single cell geometry and parameters were incorporated in the fuel cell model and calculations of the flow, concentration and current density distributions have been made. The model have been used to run simulations for the MT-PEM single cell, taking into account the results for the porosity, permeability and diffusivity calculated for the electrodes. A parametric analysis was performed for the MT-PEM single cell conditions, using intended candidate materials as guide for the single fuel cell.

The membrane ionic conductivity effect was found to be very important. The effect of the electrical conductivity of the BP on the cell polarization was also quantified and found to be important, as well as the Gas Diffusion Layer electrical conductivity. The GDL porosity can also become important as the reaction rate becomes larger. The Anode/Cathode gas inlet velocity effect was also not as significant, for the cases investigated, as the catalyst activity, which had pronounced effect on the cell performance.

The effect of different cooling fluid flow rates was investigated for a MT fuel cell, and results were found to be in agreement with experimental ones.

At the final part of the project, following the roadmap (Figure 25) from phase 1 (LT PEM structures) to phase 2 (MT PEM using LT PEM structures) to phase 3 (MT PEM with optimised structures), and after a series of different geometry GC scenarios studied with simulations, an improved GC geometry of an MT PEM was designed. The whole geometry of the optimized gas channels structures was successfully incorporated into the simulator, and its operation at MT conditions was successfully simulated and reported back to the consortium. 3D simulations included all the main effects of the MT operation at 0.48 A cm^{-2} point. Simulations demonstrated the improvement on the homogenization of the flow field, especially at the flow areas near the outlet of the gas channels, on the optimised gas channel structures, compared to the initial ones.

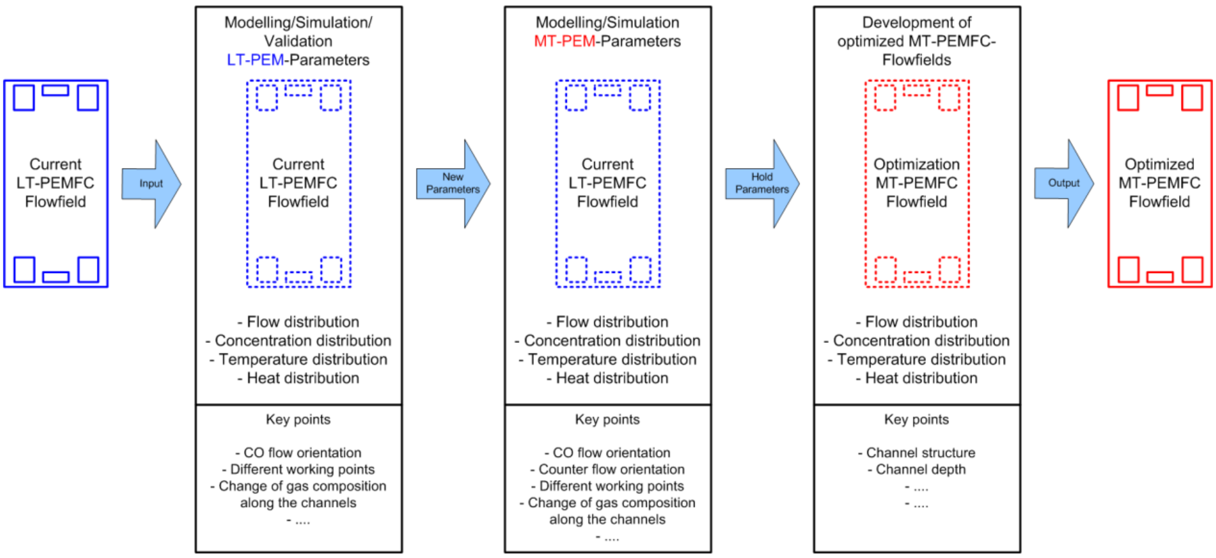


Figure 25. Roadmap for development and validation of single LT/MT-PEM fuel cell model

The used material MT-PEM properties and MT-PEM operating conditions for Phase 2 are listed in Table 1. Most known material properties were given from partners CEA and

Eisenhuth. Material properties which were not determined by the partners were taken from the international literature.

Table 1. MT-PEM properties and conditions

Material properties (MT)	
Porosity GDL/CL	0.75/0.70
Electrolyte volume fraction in CL anode/ cathode	0.51/ 0.37
Permeability GDL/CL	6×10^{-14}/ 6×10^{-14} [m^2]
Electric conductivity GDL in-/through plane	8000/200 [S/m]
Electric conductivity CL in-/through plane	8000/200 [S/m]
Electric conductivity BP in-/through plane	14780/2600 [S/m]
Electrolyte conductivity MEM	7.14 [S/m]
Electrolyte conductivity CL anode/cathode (Bruggeman Effective conductivity)	2.63 /1.64 [S/m]
Reference exchange current density anode/cathode	1.45×10^{-2}/ 1.45×10^{-7}[A $cm^{-2}{}_{MEA}$]
Thermal conductivity GDL-CL/MEM/BP	1.70/0.95/20 [W/m/K]
Volumetric heat capacity of GDL/CL/MEM/BP	0.568/1.69/1.65/1.57 [10^6 J/m^3/K]
Operating conditions (MT)	
Anode/Cathode velocities	2.65/6.04 [m/s]
Inlet Anode/Cathode	H_2+H_2O+CO_2/O_2+H_2O+N_2
Inlet mass fractions $H_{2(A)}$ /$CO_{2(A)}$/$H_2O_{(C)}$/$O_{2(C)}$	0.058/0.426/0.188/0.189
Temperature	85°C
Reference Pressure	101 [kPa]
Cell Voltage	0.65 - 0.95 [V]
Co-flow operation	

1.4 WP5

The goal of EURECA WP5 was the development of a MT-PEM µ-CHP system based on the system platform of inhouse. The main tasks were related to design issues and components developments to operate at MT-PEM Stack conditions. The work package also incorporated performance testing and degradation measurement. The adaption was guided by simulating system performance with a specific static model and intense pre-testing of developed balance of plant concepts and components. These preliminary works lead to the development, construction and testing of a MT-PEM System with the MT-PEM Full Stack from WP4.

1.4.1 System Model for a MT-PEM Fuel Cell µ-CHP System

During the reporting period several simulation were performed by using the developed static system model for a MT-PEM CHP system including the energy conversion path (Figure 26) and measurement data obtained especially from stack testing within the project. These modelling results composed the roadmap for development/ adaption of the MT-PEM system. Special focus was taken on the power electronics efficiency and the comparison of the model with data obtained from system performance evaluation.

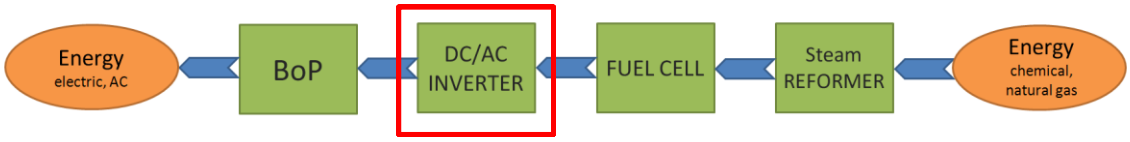


Figure 26. Energy conversion scheme of static system model

Based on measurements of the power uptake of the balance of Plant components and U/I-curves from the 7th EURECA Stack, calculations were done to show the potential of the MT-PEM-system. Figure 27 compares the electrical efficiencies of two MT-PEM System configurations:

- with the reference MEA from WP2,
- the U/I-curve of the final EURECA stack (7th stack) extrapolated to a 83 cell Full stack, and the estimated and measured BoP power uptake as well.

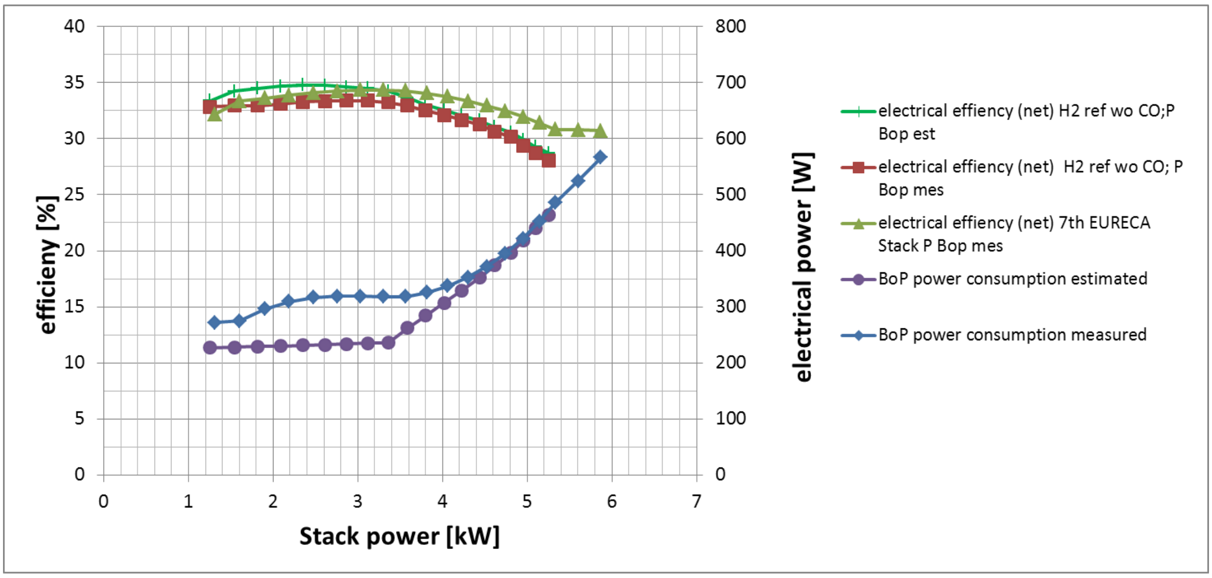


Figure 27. Calculated efficiencies with 7th Eureca Stack U/I curve and measured BoP power uptake

The maximum efficiency that is calculated with the extrapolated U/I curve of the 7th EURECA stack and the measured BoP power uptake is 34.3 %. As shown in Figure 27 the values of the BoP power uptake, extrapolated from the measured data from the MT-PEM system in

Task 5.3 show good correspondences with the primary estimated values in the upper power region. The deviations in the middle and lower power region of the stack can be partly explained with a higher base BoP consumption than expected, caused by control and sensor electronics (approx. 50 W). Figure 27 shows clearly that the goals of MS 5.1 are reachable, when the performance of the 7th Eureca Short Stack is transferred to MT-PEM system level.

1.4.2 Adaption of µ-CHP system

The main topics for the adaption of the µ-CHP system that were addressed, included the evaluation and long term testing of materials and components for the cooling circuit for the MT-PEM system with the chosen synthetic cooling fluid, followed by concept evaluation and layout of the cooling circuit for the MT-PEM system. Beside other activities the detailed engineering of the MT-PEM µ-CHP system was conducted where a new safety concept for reformer module combustion and necessary modification regarding the input of natural gas type into the µ-CHP system was realized.

The MT-PEM µ-CHP system consists of three separate modules and a socket. The modules are the Control & power electronics, the Fuel Cell Module and the Reformer Module.

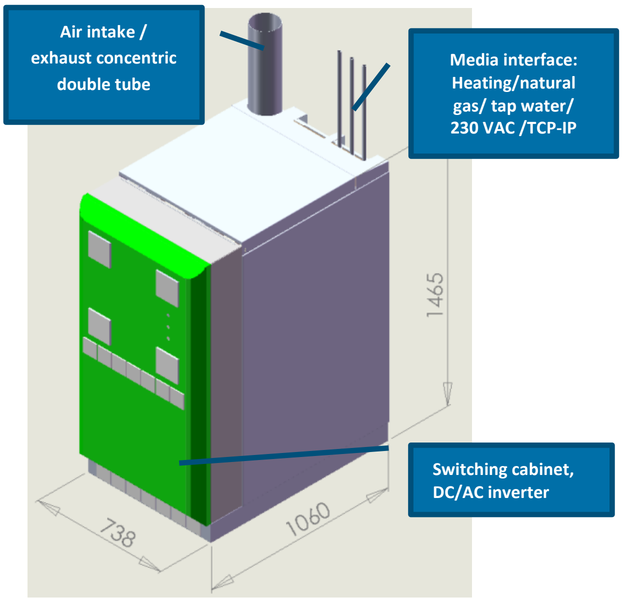


Figure 28. Detailed engineering of µ-CHP system

The installation is done by mounting the separate modules onto the socket at the installation site. Exemplary shown in Figure 29 is the Fuel Cell Module with integrated MT-PEM-Stack.

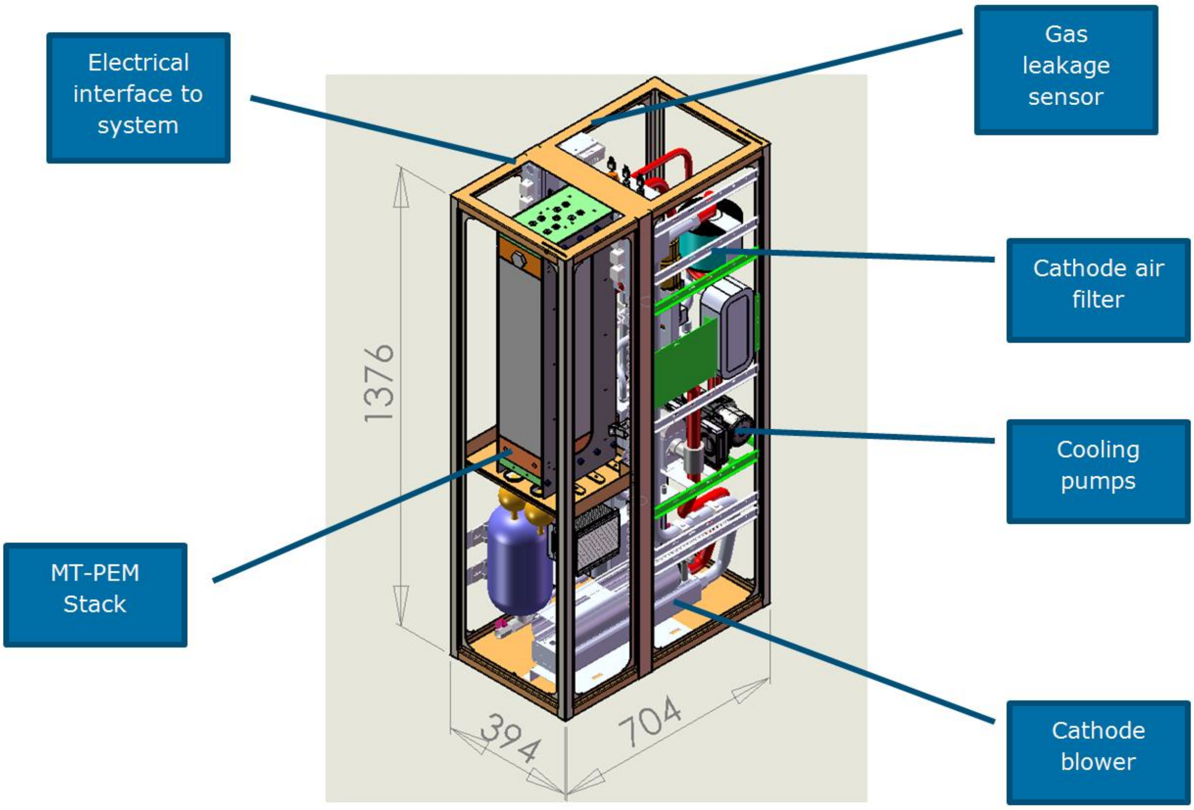


Figure 29. Detailed engineering of Fuel Cell Module

The reformer module for the MT-PEM prototype system was tested for gas conversion quality, operation parameters and long term stability prior to the integration with the MT-PEM fuel cell module and Stack. Different routines for cold start and warm start were developed and tested. At cold start conditions, the overall start time is less than 90 min, including a Stack preheating procedure.

Referring to the existing standards for fuel cell µCHP Systems namely the DIN EN 50465:2015-06 a simpler and more cost effective safety concept relating to CO emission, unburned fuel and continuous operation could be developed and successfully tested within the EURECA system. All safety critical components were identified and the related signals were analyzed regarding failure modes and the related behaviour of the µCHP system. The developed safety concept consist of hardware components such as pressure switches and safety electronics that fulfil the requirements of the existing safety standards and of software routines that supervise the proper functioning of the components by counter checking with related system parameters.

Also the gas composition of the reformer output / stack anode feed gas was measured. As expected the hydrogen content is lower when type natural gas Type L available in North Germany with lower calorific value (H_s=9,8kWh/m³, CH_4=82 Mol.-%, N_2=14 Mol.-%) is reformed. From Figure 30 it can be seen that the deviation in hydrogen content of Type L reformat is constant from Type H available in Middle and South Germany (higher calorific value, H_s=11,2kWh/m³, CH_4=97 Mol.-%, N_2=0,1 Mol.-%) reformat over the working range of the reformer.

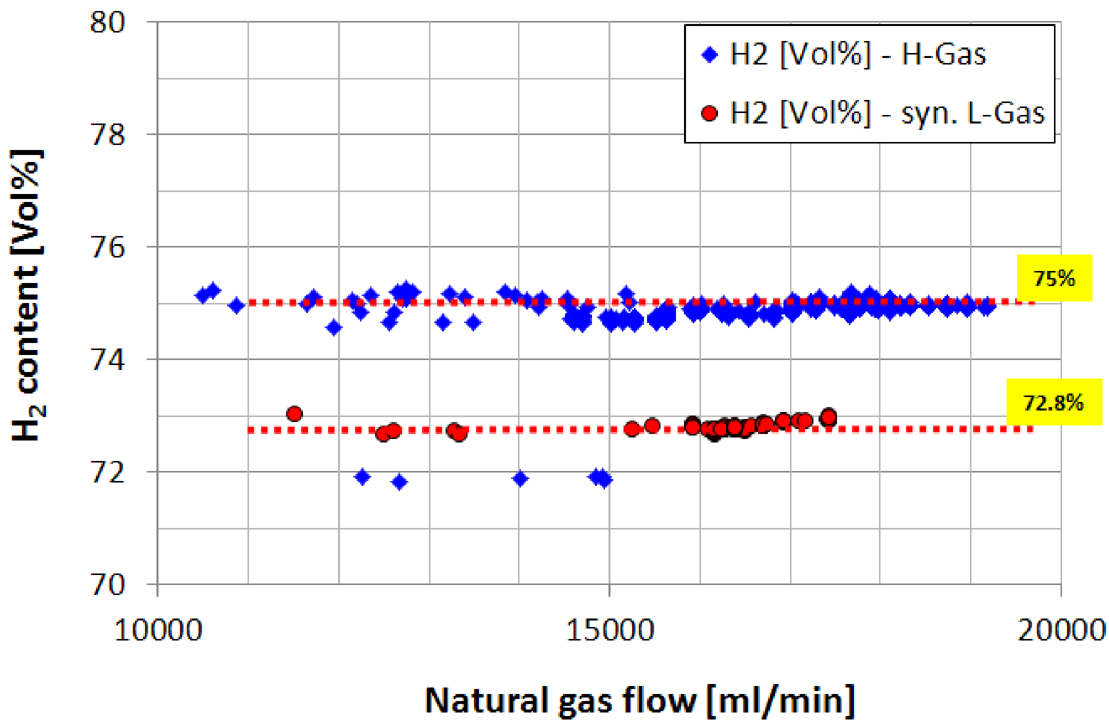


Figure 30. Hydrogen content of reformate with type H and type L natural gas

The deviation in hydrogen content was determined to be about -2.2 %. Accordingly the operating routines of the reformer with in the CHP were adapted to compensate the lowered hydrogen content for anode feed gas supply to the MT-PEM Stack.

1.4.3 Assembling, testing and optimization of prototype of the MT-PEM-FC system

The MT-PEM FC system was assembled by building and pre testing the three modules separately. The testing included checks on leak tightness, electrical wiring, sensor accuracy and a dry test of every electrical component. Afterwards the modules were assembled to the complete system and put on INHOUSE test bench. Figure 31 shows the MT-PEM system with the MT-PEM Stack on the INHOUSE test bench before it was delivered to NEXT ENERGY.

Figure 31. MT-PEM system with MT-PEM Stack

Figure 32 show data from the MT-PEM Full Stack operated in the system with natural gas type H.

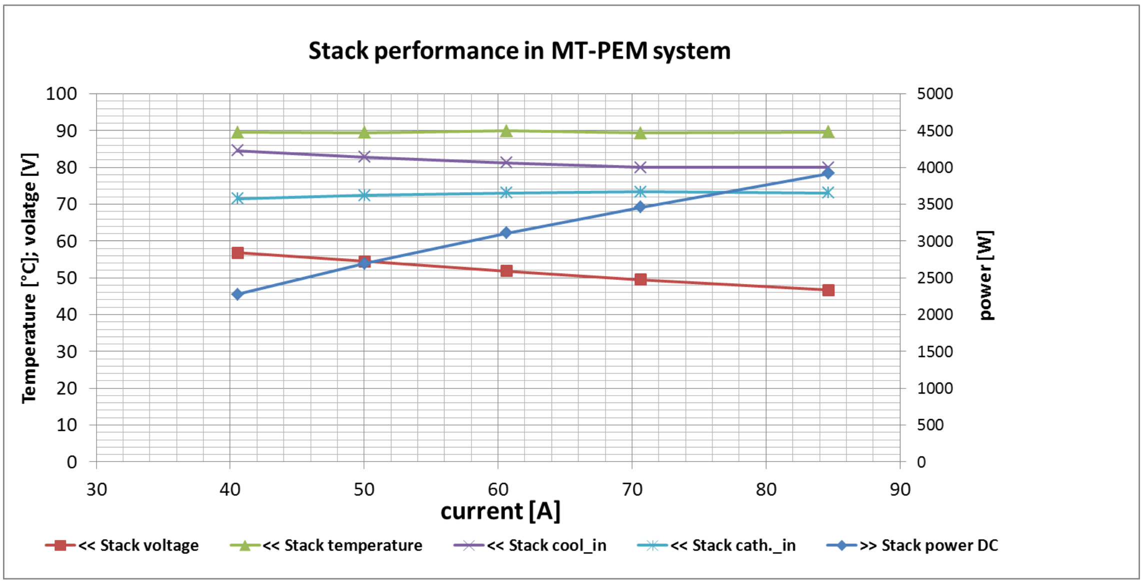


Figure 32. Stack voltage and power with corresponding operation temperatures and using of natural gas type H

After completion of the initial testing, the system was transferred to NEXT ENERGY. There it was integrated within a specialized CHP test bench and was operated by INHOUSE and NEXT ENERGY. Although the system was tested with a synthetic type L natural gas it was necessary to modify operation parameters to adapt it to the real type L natural gas available at NEXT. During the operation time measures were taken to improve and stabilise stack performance. Figure 33 shows the MT-PEM system with the MT-PEM Stack on the CHP test bench after its integration at NEXT.

Figure 33. System test stand at NEXT (left) and connected MT-PEM system (right)

1.5 WP6

In a first step, the EURECA partners analysed cost structure of the current system and evaluated the cost reduction potential for this specific type of fuel cell. A summary of the components has been set up and all project partners contributed with cost information, such as material, fabrication charges and available cost-volume information. As described in the section above there are considerable dependencies between different components within the fuel cell system. Consequently, not every cost saving of a single part contributes to overall cost reduction. The partners identified the most important interactions and identified critical cost parameters. Target of EURECA system design is a low overall cost of the fuel cell system. As starting point, cost structure of the current system without improvements from EURECA was analysed.

Within the first half of the EURECA project the partners analysed cost structures and collected data to determine further cost reduction in the case of technical improvements from EURECA and for a production volume of 1000 fuel cell systems per year. The coordinator collected cost information of all partners, in particular industrial partners, and summarized them. For confidentiality reasons these cost details were structured by the coordinator into the sections stack, MEA, Pt, reformer, BoP and others and then circulated to all partners.

The current cost information of the EURECA system for 1000 pcs/a is considered as an extrapolation. It shall be updated based on the technical results achieved in EURECA and possible changes in cost/volume information regarding components. Changes in price/volume data for components and precious metal prices on world market will be taken into account.

1.5.1 Exploitation

1.5.1.1 Generell

In the coming years and decades Europe has to face an enormous structural change in the worldwide energy supply into a decentralised, intelligent, structure based on demand and supply. Combined heat and power (CHP) will play a prominent role as a conversion technology along with solar, wind and biomass as renewable energies to fulfil the energy demand of the future. The market for combined heat and power plants (CHPs) will therefore continue to grow and become of increasing importance in case of the overall energy efficiency and the positive carbon footprint by use of the technology. While engine-driven systems are mainly used at present due to their availability, these will be replaced in future by micro CHPs based on fuel cells because of their discussed and shown advantages. Such Fuel cell CHPs are characterised by:

- High electrical and overall efficiency
- Usability of a large modulation range
- Possible low maintenance costs with a small number of moving parts
- Very low emissions (waste gas and noise)
- And possible long service intervals and overall runtime.

The separation between energy supply for transportation and for the stationary sector will become less and less distinct. In future, vehicles powered by fuel cells and batteries will function as mobile storage for the electricity grid. Hydrogen generated from wind energy will be converted by fuel cell CHPs to supply households and electric vehicles. Development of a hydrogen infrastructure for the mobile sector inevitably means development in the stationary sector. The two pathways are mutually dependent and will merge to an increasing degree. Hydrogen fuel cell systems will become increasingly attractive due to further improvements in performance values.

This development is already underway. Large automobile manufacturers such as Toyota, Honda, Hyundai and Daimler are starting with fleet trials for fuel cell vehicles in small-volume series production. In addition, the most recent developments in the stationary sector also show that stakeholders recognise the high potential of fuel cell CHPs and that the system will be soon or are ready for the market.

Global leaders for market introduction are Japan and Germany. Japan is going a straight way with focusing on small CHP system up to 1 kW electrical power. Seeing the impressive growing sales numbers for the systems within the last 5 years, the market introduction was successfully implemented in Japan and cost reduction progress could show by reducing the public funding during ramp up production to reach the factors to use the economy of scale.

With the complementary application of CHP systems for single houses and easier building integration the potential to address the European market is equal or even higher than in Japan. From the market preview the EURECA project identify an also large potential to address the energy demand of multi-family houses and small industrial applications to implement the EURECA project achievements to save cost per installed kW and to share peak load and avoid individual effects of the users.

1.5.1.2 Market potential in Europe

Within the study "Klassifizierung des Gebäudemarktes für Brennstoffzellensysteme" from INECS GmbH (July 2011) it was tried to analyse the overall market potential for fuel cell CHP systems with 5 kW electrical power in the European Union.

Based on the demographic development in the European Union, the development of the persons per household and data from Eurostat the study estimated the building structure with in the EU categorized with multifamily homes (three and more households), single family homes and ,semi-detached houses.

Overall there are approx. 207 million flats with the EU. The structure is dominated by the big member states like Germany, France, Italy, Poland, Spain and UK. For example Germany has nearly 40 million flats and approx. 21.5 million flats are located in multifamily homes. This underlines the fact why Germany is a key market but it also demonstrates that there is a quite bigger potential in EU.

60 % of the flats in the EU are located within single or semi-detached homes and 40 % in multifamily homes. That implies 82 million flats located in multifamily homes which are in principal suitable for CHP based energy supply (5 kW electrical power). But Table 15 also shows that nearly 61.5 % of all flats are located in buildings build up before 1975. This fact underlines the following hypothesis:

- Beside the installation/integration of fuel cell based CHP system in new buildings a major task is the replacement of old heating systems.
- Replacing older heating systems (build up before the eighties) implies heating systems with supply temperatures clearly above 50 °C. This replacement cannot be done by a Low Temperature PEM CHP system, which can deliver a maximum supply temperature of 50 °C and a lot of old building cannot be reconstructed in that way that a Low Temperature-PEM (LT-PEM) CHP system can be used. A Medium Temperature-PEM (MT-PEM) CHP system is able to deliver quite higher supply

temperatures and are therefore a predestined solution for an energetic reconstruction of old buildings.

Taking these two issues into account the MT-PEM clearly enhances the number of application for use of a PEM-fuel cell based CHP system.

Based on that numbers and influenced by the heating demand for hot drinking water an overall potential for PEM fuel cell based CHP systems within the EU was estimated. Two cases were taken into consideration for this estimation. The first case is an economical use of hot drinking water (20 l per person and day) and the second case is a more or less carefree case (40 l per person and day).

In absolute numbers this will be a potential market of 2.4 up to 4.9 million units or 12 GW up to 24 GW.

1.5.1.3 Economical Chances of success

The potential for economic success can be significant. For the industrial partners of the project, there are the following potential economic possibilities after finishing EURECA-project in 2015; the following one till two years will be needed for upgrading MT-PEM Fuel Cell from a prototype to a final product. In addition, new partnerships for commercialization of the Fuel Cell system have to establish in this period.

Therefore, for 2016 and 2017, we expect a slightly positive impact. If the final product is realised, a possible entrance into the market from 2018 / 2019 on, can be realistic and positive impacts on the performance of the industrial partners are expected.

1.5.1.4 Market design and market share of the industrial Partners

Today the industrial partners are significant players in the Fuel cell industry. INHOUSE is one of the significant actors in the field of domestic heating applications. EISENHUTH is one of two guiding companies in the field of delivering graphite bipolar plates for use in various applications. CEGASA is one of the important MEA manufacturers in Spain.

The customers of these companies mainly work in the NT-PEM or HT-PEM business, but not so far in the MT-PEM area. Here is still a big potential.

The products such as the system, as well as the MEA and the bipolar plates and gaskets for MT-PEM Fuel Cell, get so far a positive echo in talks with potential customers.

They mention the two big advantages of MT-PEM Fuel Cell systems compare to LT-PEM Fuel Cell systems. On the one hand the ”EURECA system” is well prepared for using in existing heating infrastructure because of the provided heat on high temperature level. The second important aspect is the target lifetime what leads to a good economic reliability. To increase the degree of popularity of the "EURECA system", cooperation with the heating contractor association could be an effective step.

1.5.2 Scientific and Economical Connectivity

Due to the global climate warming up and due to the trend of renewable energies, it is evident that the Fuel cell will be one of the key technologies for an efficient use in the energy sector.

Together with stable political support and possible actions for a market entrance as well as the high added value of more than 80 % makes the European Fuel Cell industry competitive.

Based on stable economical frames and with actions in industrial politics in Europe, the added value can be also increased. According to studies, it is possible that only with fuel cells in stationary applications, more than 1.5 Billion Euros can be realised as potential turnover and more than 5000 people can be employed.

The result of this project, especially the successful implementation of the MT-PEM technology, can bring the industrial companies a big step forward to become a significant player in the µ-CHP market. Especially in this area, an enormous market potential is existent, but also high technological requirements, such as an operation time of 40.000 h to 50.000 h.

The MT-PEM CHP system consists of the main reformer, fuel cell module with stack and control unit modules (fig. 1). This general design will be retained for optimized MT-PEM CHP systems with internal hydrogen generation.

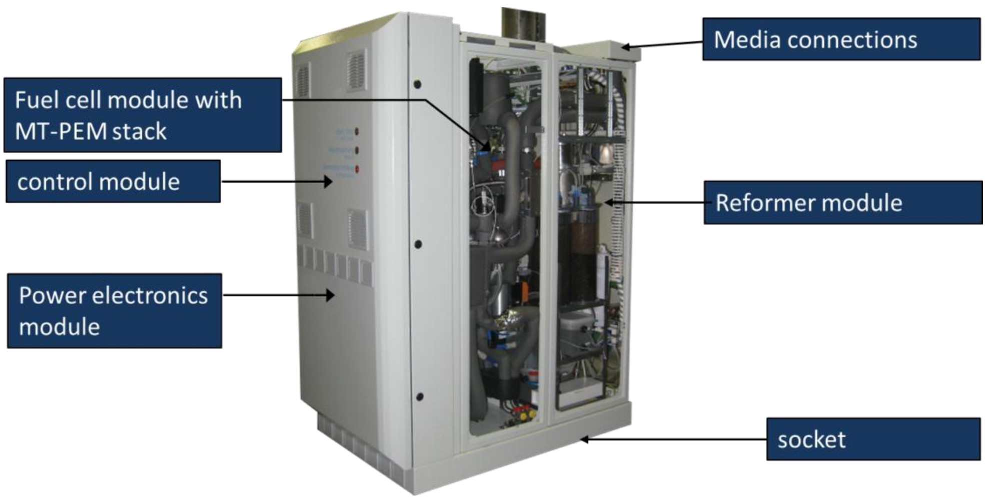


Figure 1. Modular design, overall system view MT-PEM CHP system

At the present state of development and also for the manufacture of the small-scale series, a modular production can be set up in accordance with figure 2. Components are obtained from outside specialized suppliers, according to our own design specifications. These single components are used to produce functional assembly groups which are constructed according to the modular design of the MT-PEM - CHP system and which are also best suitable for later module assembly and easy maintenance access during system operation time. These functional assembly groups are combined too system assembly groups which in turn are installed in reformer module, fuel cell module, control- and power electronics module using cellular production. Completion of the MT-PEM CHP system takes place using the same joining production. With increasing quantities a decision has to be made whether purchasing a component or in-house production is the more economical alternative in respect of the supplier parts and individual assemblies. Due to the small quantities, the supplier will not offer any cost-reduction initially, so that in-house production may provide the more economical alternative to begin with (e.g. process water treatment, containers for condensate and mixed-bed resin).

But also for increasing number of systems, the production capacity can be easily extended through paralleling the defined production process/ steps as shown in figure 2. Also a more or less high quantity production with 5000 or 10.000 systems per year will be also semi-automated process. Which means some high quantity components like MEA or BPP will be produced in a fully automated process, but system or stack assembly will be made by workers supported by tools and assembly fixtures.

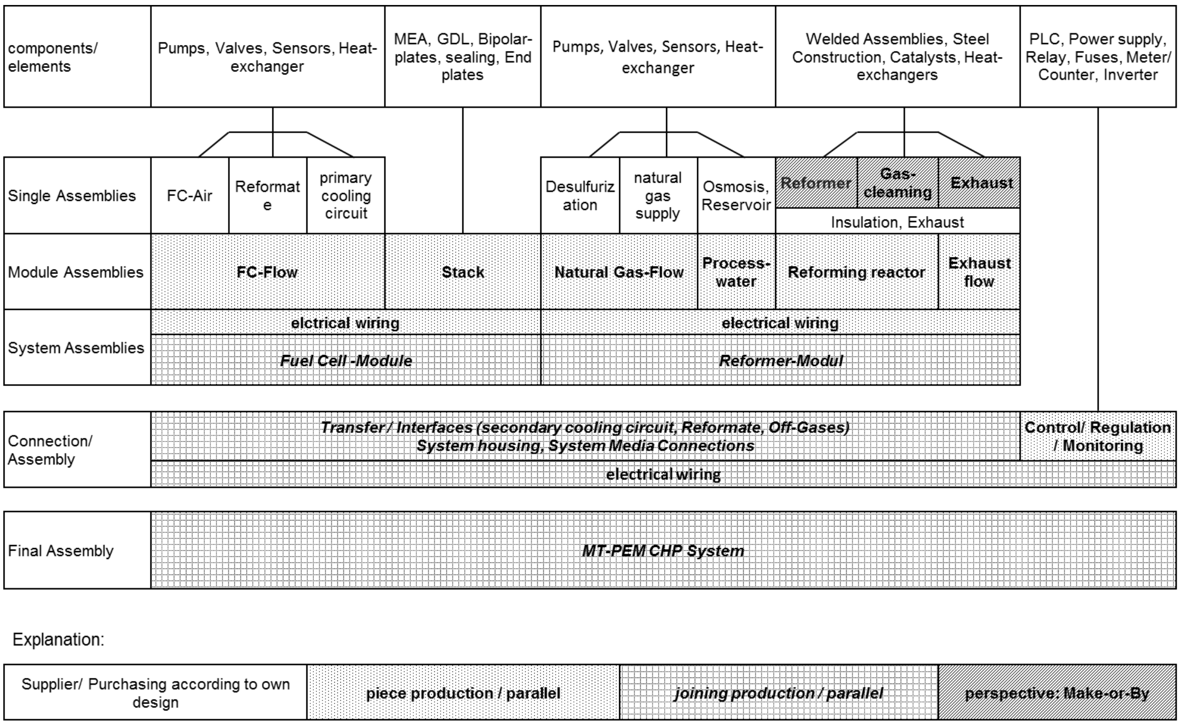


Figure 2. Possible modular production process of MT-PEM CHP system