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1. EXECUTIVE SUMMARY

A novel cross-linked polybenzimidazole membrane reinforced by an electrospun cross-linked nanofibre web has been developed and its preparation scaled-up. The membrane has proton conductivity of 130 mS/cm, an acid doping level of 21 molecules per polymer repeat unit, and Young's Modulus of 80 MPa. Membranes of surface area 400 cm^2 have been produced batch-wise and transferred to WP4 for development of full size MEAs for the HT PEMFC stack.

A multi-scale modelling tool has been developed to investigate how degradation effects during FC operation may be mitigated. The proposed confinement strategy could lead to the use of membranes with increased acid doping levels.

Non-noble metal CoS_2/C, NiS_2/C, $CoSe_2/C$ and $NiSe_2/C$ cathode electrocatalysts were developed. CoS_2 with cubic structure has the best activity towards the oxygen reduction reaction in an acidic electrolyte. A PtNi/MWCNT cathode catalyst was prepared and scaled-up. A polyol synthesis route to $Pt/WC\text{-}C\text{-}CeO_2$ anode catalyst was developed and scaled-up. Anode and cathode catalysts were transferred to WP4 for electrode preparation.

Ink composition and deposition were optimised using a commercial Pt/C catalyst. The resulting electrodes were used with the ARTEMIS cross-linked and reinforced membrane to produce initially small size (25 cm^2) and finally full size (200 cm^2) ARTEMIS MEAs, by optimising the assembly parameters and sub-gaskets.

MEA performance exceeds the target of 0.5 W/cm^2 at 1 A/cm^2 in 25 cm^2 single cell tests and is significantly higher than that of the reference commercial MEA (25 % higher at 1.2 A/cm^2). Such MEAs have been operated using the ARTEMIS range extender protocol in 1000 hour and 2000 hour longevity tests. The voltage decay is low (8 µV/h), and after 1000 h operation the power density delivered at full load is still close to 0.75 W/cm^2. Full size MEAs have been transferred to WP5 for short stack development.

Two cell plate formulations were processed and tested for performance. Mechanical tests were performed to confirm mechanical properties of this material. Seal material was chosen and the required seal thickness calculated. The hardware components design was completed.

Following preliminary performance tests on single cells to optimise the HT-PEMFC stack assembly, a four-cell high temperature PEMFC stack was assembled and tested. This stack produces >0.3 kWe at 160 °C at ambient pressure and without humidification for currents over 165 A (825 mA/cm2) and at 180 °C for currents over 140 A (700 mA/cm2). A 3 kW HT-PEMFC stack was configured using the results from this four-cell stack. It was determined that 48 cells of average performance are required to produce 3 kWe at 160 °C in the range of 125-160 A (BoL to EoL) and at 180 °C in the range of 110 A (BoL) to 140 A (EoL). Based on 15% voltage decay as end-of-life condition this configuration allows for the output of 3 kWe from BoL to EoL in a suitable current range.

Vehicle models were developed to assess the effects of an HT-PEMFC stack on the vehicle range through the simulation of different driving cycles. For a 3 kW stack, two solutions were analysed: (i) On-board generation for light commercial vehicles, where the fuel cell is used to supply power to the auxiliaries, where the vehicle runtime was shown to be improved by 11% for the NEDC, and by 19% for the ASTERICS driving cycle, with respect to the full electric vehicle; (ii) Charge-sustaining suitable for passenger cars, where the fuel cell is used to supply power for traction, where it was shown that a 3 kW fuel cell can ensure battery charging in the cases of Urban Driving Cycles and Common Artemis Driving Cycles.

A dissemination event was held, and 6 journal papers have been published or are planned.

Overall improved high temperature PEMFC MEA components and MEAs have been developed, leading to ARTEMIS HT-PEMFC technology for future research and development, and the approach of a HT PEMFC stack as a range extender has been validated.

2. SUMMARY DESCRIPTION OF PROJECT CONTEXT AND OBJECTIVES

PROJECT CONTEXT AND OBJECTIVES

There is increasing industrial interest in developing HT-PEMFC systems in conjunction with a Diesel or methanol reformer to continuously charge batteries on-board of automotive vehicles, thus extending the range to several hundred kilometres, using the existing infrastructure for hydrocarbon fuels. HT-PEMFC systems are being developed commercially for backup systems in remote areas or developing countries where a long operation time is required when the grid fails. High temperature fuel cells offer advantages for the overall system as they require less balance of plant components and offer high tolerance to CO and other pollutants. Unlike low temperature PEM fuel cells that depend on perfluorosulfonic acid type membranes, HT-PEM fuel cells need no humidification and their use therefore eliminates water management issues. However, due to the temperature range of operation (150 to 180 °C), the challenge is to improve all the materials involved in the complete stack. This challenge includes the polymer membrane that should offer excellent conduction, mechanical and thermal properties to the stack components (seals, bipolar plates and coolant). The catalyst layers and gas diffusion layers (anode and cathode), while including low catalyst content, have to be resistant to corrosion, dissolution or aggregation, which would lead to a decrease in performance or a failure of the system. The purpose of ARTEMIS was to develop and optimise alternative materials (membrane, catalysts, support, plate composite materials, seals and gaskets) for a new generation of European MEAs to be integrated into a high temperature PEMFC stack.

The objective was to base the MEAs on polybenzimidazole type membranes and improved catalyst layers providing low catalyst loading and high efficiency at high temperature as well as a high tolerance to pollutants such as CO. High temperature properties are maintained in non-pressurised cell hardware and without gas humidification. Costs are reduced since the technology avoids the use of fluorine-based chemistries characteristic of low temperature PEMFC, hydrocarbon polymer-based chemistries and corresponding membranes being significantly less costly. The MEAs were required to offer long-term stable properties under various conditions of operation relevant to the range extender application, in particular voltage cycling and start/stop. It was intended to investigate the possibility of producing HT PEMFC MEAs at pilot level. Due to their operation at 150-180 °C, HT-PEM stacks are the most appropriate PEM technology for a transportation range extender. However, it was critical not only to develop new and less expensive materials, but also to understand the failure mechanisms that occur in all the parts of the stack (membrane, stack components, catalysts and supports). The understanding of such mechanisms leads to increased durability and lifetime of the system by improved understanding of the different failure modes. The operation conditions of a fuel cell for the range extender application differ from those when the PEMFC stack is used for the power train. Thus the stack is subjected to fewer transients and operates in semi-continuous mode. But stop/start events corresponding to the normal start up and shut down of vehicle operation are present. It was important therefore to address particular attention to the definition and use of appropriate accelerated stress testing protocols. Such protocols are for use by partners to obtain a first assessment and validation of the MEAs and bipolar plates, and will feed input to the development of alternative and improved materials (characterised by in situ and ex situ methods) intended for integration into complete MEAs, and the MEAs into a stack.

3. DESCRIPTION OF THE MAIN SCIENTIFIC AND TECHNICAL RESULTS

3.1 WP2: SPECIFICATIONS, PROTOCOLS DEFINITION

OBJECTIVES

The objectives of this work package were to:

- define characterisation and testing protocols for use in WP3, WP4 and WP6.
- define relevant operating conditions for range extender fuel cell application, in situ and ex situ characterisation methods to be used for membrane/catalyst layers / MEA evaluation
- apply the defined protocols for a parametric study of the influence of working parameters on MEA durability and aging mechanisms
- select MEAs for ageing tests and stack fabrication

To achieve deliverables D2.1-2.2 in RP1

SUMMARY OF OUTPUT FROM WP2

Testing protocols have been defined to evaluate the behaviour of the PEMFC stack and its membrane electrode assemblies (MEAs) in the specific operating conditions of the range extender application in order to screen for suitable materials. Protocols intended for the selection of suitable materials comprises I-V characteristics and cell operation at constant current. Protocols comprising load cycling characterise MEAs under the range extender application. Furthermore, testing protocols to study degradation phenomena within the MEA at high temperature were defined, including parametric studies under pollutant-containing hydrogen. A number of physicochemical and electrochemical protocols were applied to gain more insight into the characteristics of MEA components (membrane and catalysts). This was essential to select the most prospective candidates.

DETAILED SUMMARY OF ACHIEVEMENTS IN WP2

Baseline fuel cell test protocols

Protocols for high temperature PEMFC were not included in the harmonization exercise that began during the course of ARTEMIS, although ARTEMIS did participate in the first meetings. In situ protocols were therefore developed to be applied to ARTEMIS MEAs in single cells and stack. In these protocols, the operating pressure was 1.1 bara at the anode and 1 bara at the cathode, cell temperature in the range 130 - 180 °C under dry hydrogen and dry air at stoichiometries of 1.5 (H_2) and 2.5 (air). The protocols included a break-in procedure at 160 °C to reach constant voltage operation at 0.2 A/cm², of duration generally not exceeding 100 h.

Protocols were developed for continuous operation designed at down-selection of project materials suitable for further investigation under cycling mode.

Cycling mode was defined in order to characterise MEAs under simulated automotive Range Extender application (auxiliary unit alimentation and battery charging). The Range Extender drive cycle differs from commonly adopted ones that charge the battery rack only when the battery charge level is below a minimum value. For ARTEMIS, it was intended that the stack would be used to power the auxiliary units (dashboard, lamps, headlights, chips, ECU etc.,) and to recharge the batteries (recharge also coupled with braking regeneration). Applied at single cell level, the cycle comprises a 60 min cycle during which the MEA is subjected to three different load demands (0.2, 0.4, 0.6 A/cm²) depending on the battery charge level. For a battery charge level below 50 %, the load demand is considered to be 0.6 A/cm² until a charge level of 60 % is achieved, and then the load demand reduced to 0.4 A/cm² until full charge is reached. While the batteries are in discharge, it was considered that the load demand should be fixed at the minimum to power the auxiliary units, Figure 2.1.

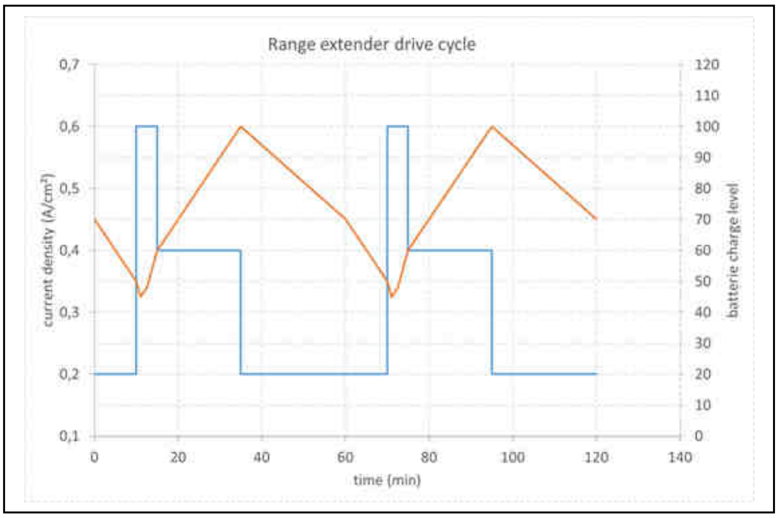


Figure 2.1: Range extender drive cycle protocol in ARTEMIS

Protocols for parametric study of the influence of working parameters on MEA durability and aging mechanisms

Parametric studies under polluted anode gas (CO, H_2S) were defined.

Ex situ characterisation protocols: membrane, catalyst layer

Ex situ characterisation protocols were established for membranes, electrocatalysts and catalyst layers. In particular, protocols for the structural characterisation (XRD), morphology (SEM, TEM) and composition (EDX) determination, as well as particle size distribution (TEM) analysis of electrocatalysts were defined. With regard to the electrochemical characterization, two protocols were defined to assess the catalytic activity of electrodes towards the reactions of interest: O_2 reduction and H_2 oxidation in rotating disk electrode (RDE) and in modified floating gas diffusion electrode setup (half-membrane electrode assembly). In addition, another protocol was specifically defined to evaluate the tolerance to carbon monoxide poisoning of those electrocatalysts containing platinum in their composition. Characterisation protocols for the membrane comprised the determination of the main parameters determining the selection of membrane materials: conductivity, acid doping level, membrane tensile strength tests and polymer solution inherent viscosity.

MEA relevant parameters criteria selection for ageing tests and stack fabrication

Only the materials that matched the criteria defined in the characterization protocols were selected for MEA preparation.

CONCLUSIONS

- Ex situ characterisation protocols for physicochemical and electrochemical characterisation were described to allow selection of the most appropriate membrane, catalyst and plate materials for HT PEMFC
- The evaluation of the behaviour of HT PEMFC MEAs and stack under the specific operating conditions of the range extender application as an auxiliary power unit and for battery charging required the definition of specific testing protocols, including continuous operation and load cycling,
- The load cycling mode protocols comprised a drive cycle of duration 60 minutes and load demands of 0.2, 0.4 and 0.6 A cm^{-2} depending on the battery charge level.
- Protocols for cell operation in the presence of contaminated (CO, H_2S) anode feed were developed.

3.2 WP3: MEMBRANE AND ELECTROCATALYST DEVELOPMENT AND MODELLING

OBJECTIVES

Work package 3 includes both experimental and modelling work on materials development for high temperature acid doped proton conducting membranes and anode and cathode catalysts. It included:

- Development of durable and mechanically stable membranes for operation up to 180 °C;
- Anode and cathode catalysts and their supports with improved tolerance to H2S and CO and phosphoric acid;
- Upscale of the final membrane formulation and membrane casting to the amounts required for ARTEMIS MEAs and stack;
- Upscale of catalysts;
- Modelling at the level of the complete electrode

To achieve deliverables D3.1-3.10 in RP1 and contribute to MS1 in RP1 and MS3 in RP2.

SUMMARY OF OUTPUT FROM WP3

A novel membrane based on a stabilised and reinforced polybenzimidazole (PBI)/phosphoric acid (PA) system has been successfully developed and up-scaled. Reinforcement and cross-linking of the PBI/PA system increase the mechanical resistance of the membrane without sacrificing the acid doping level and the proton conductivity. The membrane is characterised by an acid doping level of 21 PA molecules per polymer repeat unit, proton conductivity 130 mS/cm and Young's modulus of 80 MPa. An anode catalyst based on platinum dispersed on a support prepared from tungsten carbide doped with cerium oxide has been developed. Pt supported on WC-C-CeO_2 show better behaviour than those supported on WC(commercial)-CeO_2 and has higher CO tolerance than Pt/C. Carbon-supported iron/cobalt, nickel/cobalt and platinum/nickel cathode catalysts have been prepared, and the Pt-Ni/C catalyst down-selected for MEA preparation on the basis of ex situ characteristics.

DETAILED SUMMARY OF ACHIEVEMENTS IN WP3

Membrane Development

During HT-PEMFC operation, the temperature and compressive stress applied to the membrane leads to viscoelastic flow of the material. This phenomenon results in the thinning and creeping of the membrane. Four main strategies are known to be relevant to face the thinning/creeping of the membrane:

1. Use of a cross-linked polymer
2. Increase of the polymer solid content within the membrane:
   Increase the PBI concentration of the polymer solution
   Integration of a PBI-based reinforcement material
3. Increase the length of the polymer chain (higher Mw)
4. Decrease of the polymer chain flexibility (use of para-PBI)

In RP1 CNRS developed a cross-linked highly acid doped polybenzimidazole (PBI) membrane that exceeded the target conductivity at the temperature of operation. In RP2 this membrane has been further stabilised by incorporation of a cross-linked electrospun reinforcement, while maintaining a high Acid Doping Level (ADL) and high proton conductivity.

The route developed at CNRS builds upon the specific solubility properties of PBI in a mixture of phosphoric (PA) and polyphosphoric acid (PPA) and is based on a combination of a themoreversible sol-gel transition, in situ cross-linking, and integration of cross-linked PBI-based nanofibres. The phase behaviour of the system

PBI/PPA/PA, as a function of temperature and PPA concentration, is schematically represented in Figure 3.1. At HT-PEMFC operating temperature (130 - 200 °C) and total or partial hydrolysis of PPA, the PBI/PPA/PA system is (or can be) a solution. A cross-linking step, following the dissolution process, allows the PBI/PA gel domain to be enlarged by preventing the re-dissolution of the membrane at all temperatures and PPA/PA compositions. The cross-linking reaction is initiated by the direct incorporation of a cross-linker agent and compatibiliser into the PBI/PPA solution. The solution is cast on a pre-treated glass plate with a mechanized doctor blade. At this stage, the reinforcement material is integrated into the cast polymer solution.

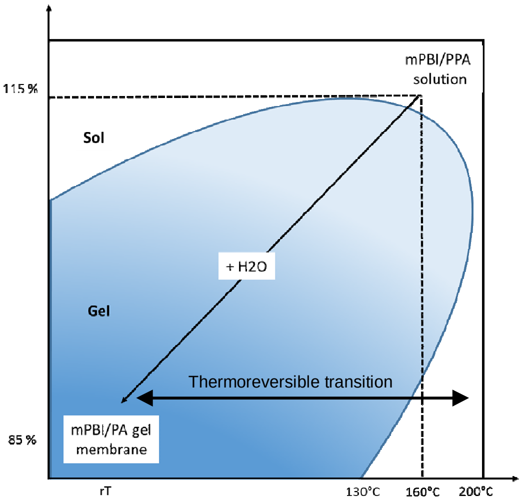


Figure 3.1: Thermoreversible sol-gel transition of the PBI/PPA/PA system

The reinforcement material comprises a 10 µm mat of cross-linked PBI-nanofibres prepared by electrospinning, Figure 3.2 (left).

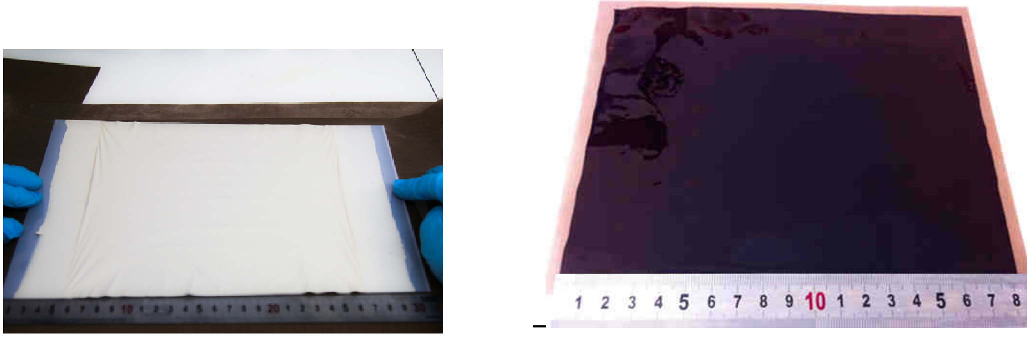


Figure 3.2: Electrospun cross-linked nanofibre reinforcement (left) and acid-doped, cross-linked and reinforced PBI membrane (right).

The reinforcement material is placed on the top of freshly cast PBI/PPA/PA solution and the whole system thermally cured to complete the cross-linking reaction and remove the compatibiliser, and then cooled to room temperature. The final dimension of the membrane (thickness and surface area) depends on the geometrical parameters during casting and the dimension of the reinforcement material. The acid-doped, reinforced and cross-linked membrane (Figure 3.2) can be easily removed from its support. In the final optimization step for membrane preparation, higher molecular weight PBI characterized by an inherent viscosity of 1.1 dL/g was used to provide additional mechanical strength.

Membrane characterisation

25 and 60 µm thickness membranes were prepared using different doctor blades. Scanning electron microscopy (SEM) shows the 10 µm thickness reinforcement material to be situated approximately equidistantly from the top and bottom surfaces of the membrane in each case, Figure 3.3.

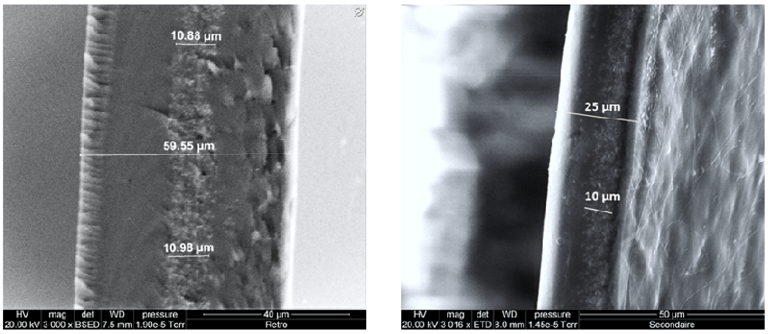


Figure 3.3: SEM micrograph of cross-linked PBI nanofibre reinforced PBI membranes

Figure 3.4 shows the acid doping level and conductivity of the reference and ARTEMIS membranes.

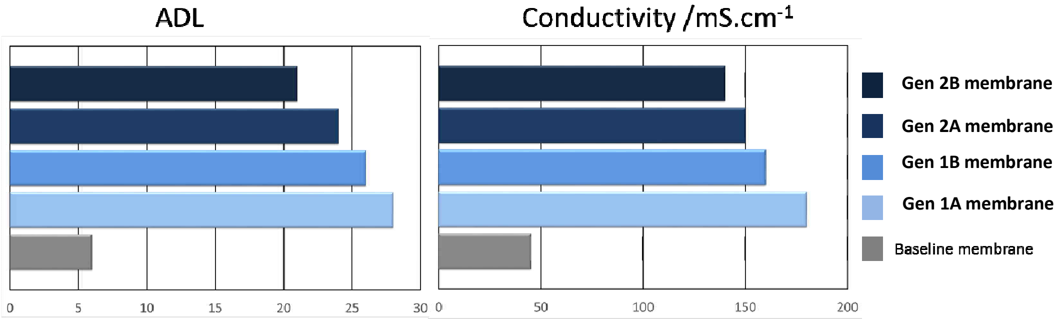


Figure 3.4: ADL and conductivity of reference and ARTEMIS membranes

Typical ADL and conductivity of ARTEMIS membranes are 28-21 moles PA/polymer repeat unit and 180-140 mS/cm, respectively, which are significantly higher than those of the reference membrane, 6 moles PA/polymer repeat unit and 45 mS/cm respectively. The Young's modulus, stress at break and strain at break of the reference and two formulations of the ARTEMIS membrane (early and final) are provided in Table 3.1.

Table 3.1: Young's modulus, stress at break and strain at break of reference and ARTEMIS membranes

	Young modulus (MPa)	Stress at break (MPa)	Strain at break (%)
Reference membrane	70	26	160
ARTEMIS membrane-1	5	2	45
ARTEMIS membrane-2	80	5	40

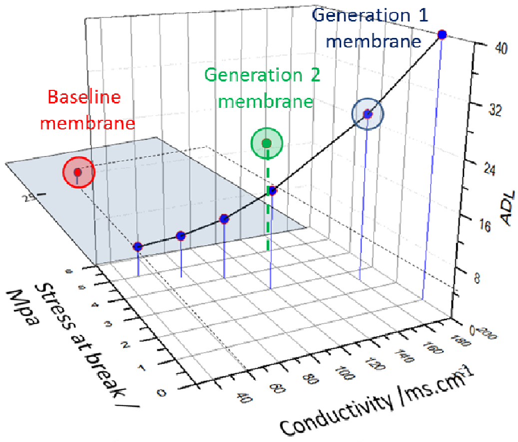


Figure 3.5: Stress at break, conductivity and ADL of reference, ARTEMIS–1 and ARTEMIS–2 membranes

The Young's modulus of the ARTEMIS-2 membrane is higher than that of the reference, although its stress and break and strain at break are lower. The salient properties of the ARTEMIS-2 membrane compared to reference and ARTEMIS-1 are summarised by Figure 3.5. This representation shows that the ARTEMIS-2 membrane breaks the trend of associating high ADL and high conductivity with poor mechanical properties, which was the objective of the work of Task 3.1.

Catalyst development

Anode catalysts: CeO_2-doped WC-C support was prepared by hydrolysis deposition of CeO_2 in WC-C dispersion, followed by thermal treatment. WC-C was obtained by solid phase synthesis. Pt (polyol methods) was dispersed on WC-C-CeO_2 at 30 and 50 wt. % nominal metal loadings. Table 3.2 shows the main properties of the anode catalysts synthesised.

Table 3.2: Characteristics of ARTEMIS anode catalysts

Catalyst	Synthesis method	Pt nominal / wt. %	Pt real / wt. %	Crystallite size[a] / nm
Pt/Vulcan® XC72R	“polyol I”	50	41	2.0±0.2
Pt/Vulcan® XC72R	“polyol II”	50	53	2.8±0.2
Pt/WC-C-CeO_2	“polyol II”	50	50	3.7
Pt/WC-C-CeO_2	“polyol II”	30	34	2.8±0.2

Cathode catalysts: The support consisted of multiwall carbon nanotubes (C), which were functionalised by chemical oxidation, and characterized by TPD and textural analysis. The selected functionalisation procedure guaranteed good textural properties and had a minimum effect upon their structure and mass loss. The catalysts M_xX_y/C (M= Co, Ni; M´ = Fe) and Pt-Ni/C (50 wt. % nominal metal, Pt:Ni atomic ratio 2:1) were prepared by different conventional chemical methods. X-ray diffraction allowed estimation of the PtNi crystallite size as 3.1 ± 0.4 nm, and EDAX analysis provided the atomic ratio between Pt:Ni. TEM analysis indicates a high dispersion of Pt/Ni on the support.

Catalyst ex situ characterisation

Electrochemical characterisation of Pt/WC-C-CeO_2 (34 wt. %) and Pt/WC-CeO_2 (29 wt. %) catalysts, towards the H_2 and H_2/CO oxidation in RDE configuration was performed at 20 °C. The electrolyte was saturated either with H_2 or H_2/CO/CO_2 (78 % vol., 5000 ppm CO, 21.5 % vol. CO_2). Figure 3.6 shows *j-E* curves for both catalysts.

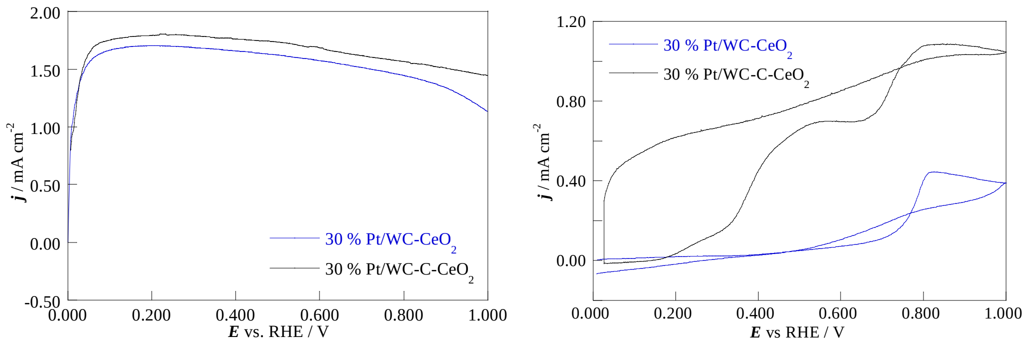


Figure 3.6: j-E curves of Pt based catalysts towards H_2 and H_2/CO/CO_2 oxidation in H_2SO_4 0.50 M (i, H_2 and H_2/CO/CO_2 adsorption at 0.015 V vs. OCP for 20 min. ii, LSV at 20 mV s^{-1}, 2500 rpm).

Pt nanoparticles supported on WC-C-CeO_2 show better behaviour than those supported on WC (commercial)-CeO_2. Figure 3.7(a) depicts the CO electro-oxidation curves for Pt/WC-C-CeO_2 and Pt/C commercial catalysts and shows the improved tolerance to CO for the former ARTEMIS support, compared with a commercial material. The *j-E* curves in Figure 3.7(b) correspond to H_2 and H_2/CO/CO_2 oxidation in hot concentrated H_3PO_4 for Pt/WC-C-CeO_2 (50 % wt.) in gas diffusion electrode.

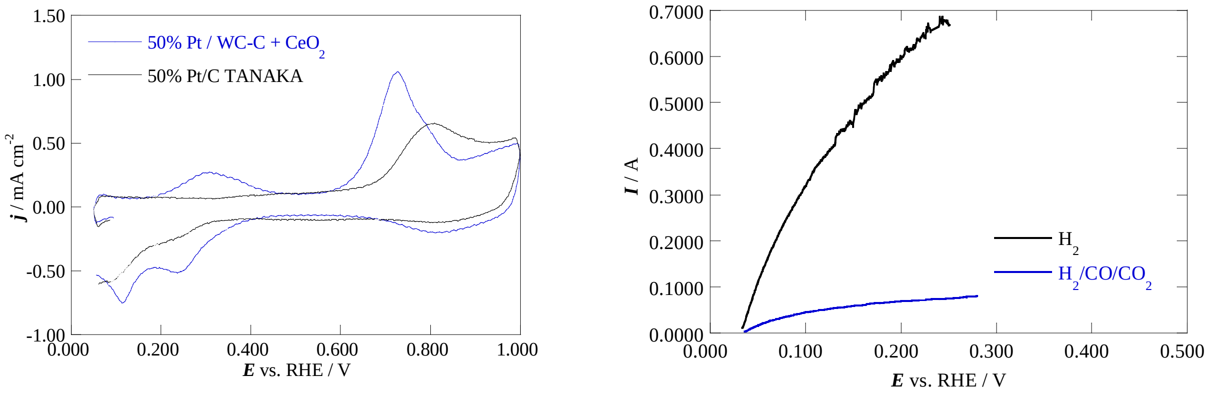


Figure 3.7: (a) Left: Electrooxidation of CO on Pt/WC-C-CeO_2 (50 % wt.) and Pt/C (50 % wt.) in H_2SO_4 0.50 M at room temperature (b) Right: j-E curves of Pt/WC-C-CeO_2 (50 % wt.) catalyzed GDE fed with H_2 and H_2/CO/CO_2 in H_3PO_4 89 % wt. at 140 °C.

Cathode catalysts were electrochemically characterised in RDE configuration, using liquid electrolyte ($HClO_4$ 0.1 M and H_3PO_4 15 M), and in three-electrode cell and half-MEA configuration as well. All catalysts were active towards O_2 reduction reaction. The FeCo/C catalysed gas diffusion electrode (three-electrode cell with H_3PO_4 89 wt.% at 160 °C), showed a decrease in the OCP, while the OCP is constant in the case of Pt-Ni/C catalyst, which was therefore chosen for upscale and implementation in MEAs.

Membrane and catalyst stability - degradation mechanisms and their mitigation

The membrane stability in situ was assessed by continuous fuel cell operation in a 25 cm² MEA. The voltage decay over 1000 hours was 8 µV/h, Figure 3.8.

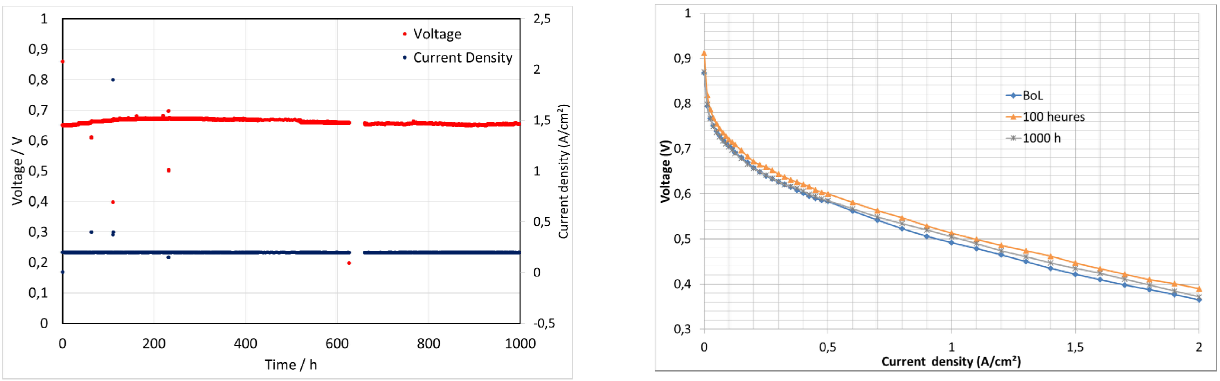


Figure 3.8: I/V characteristics of A3817 MEA comprising the ARTEMIS – 2 membrane. Anode: commercial, cathode: ARTEMIS cathode with commercial catalyst. 160 °C, H2/air, 1 bara, 1.5/2.5, no humidification.

With regard to degradation mechanisms in HT-PEMFC, focus was made on phosphoric acid loss caused by gas dynamic stress and acid leaching by water and reagents crossover through the membrane caused by peroxide chemical attacks of the membrane. An optimal cathode catalyst layer (CL) configuration with regards to catalyst distribution was proposed to effectively reduce gas dynamic stresses and production of water and peroxide radicals at the CL/membrane interface. At the pore-scale, a morphological model of CL was modelled and it was found that the proper catalyst particle redistribution reduces fluid dynamic stresses by an order of magnitude. At the macro-scale, a 3-D model of the MEA was developed with cathode CL of several sub-layers with different catalyst loadings. The adopted macroscopic model predicts stress reduction up to 69% using a feasible mitigation strategy, with only 9% efficiency reduction at high current densities.

In the macroscopic modelling, a smooth profile of a given catalyst particle distribution within the cathode CL can be obtained by a large number of subdivisions of CCL. Since this is clearly unpractical for manufacturing purposes, a given analytic distribution function was substituted with a step-wise approximation presenting a quasi-homogeneous dispersion of Pt in each sub-layer. Four different configurations of CCL were selected:

- **homo**: The catalyst is homogeneously distributed within the anode and the cathode CL (conventional);
- **case1**: The catalyst is homogeneously distributed within the anode CL while it presents an exponentially decaying distribution (towards the membrane) within the cathode CL, which is made by piling up three sub-layers with equal thickness.
- **case2**: The catalyst is homogeneously distributed within the anode CL while it presents an exponentially decaying distribution (towards the membrane) within the cathode CL, which is made by piling up three sub-layers with the same amount of catalyst metal mixed with different amount carbon support and binder.
- **case3**: same as **case1**, but the cathode CL is made by piling up five sub-layers with equal thickness.

In macro-scale, this reduction is about 40%, 12% and 69% for case1, case2 and case3, respectively. By means of the comparison between two approaches of CL fabrication (case1 and case2), with regards to gas dynamic stress reduction, the fabrication approach with the same thickness is preferable. The same reduction has been observed for water flux at the cathode CL/membrane interface. Results from the pore-scale simulation can be considered as an upper limit for macroscopic results due to a simplified treatment of the CL. In fact, comparison between the CL discretization by three (case1) and five (case3) sub-layers shows this trend, namely larger number of subdivisions smaller fluid dynamic stresses.

However, this increase in discretization number negatively affects the overall performance of fuel cell. Additional details on how the flow field is affected by the catalyst distribution with exponential decay towards the membrane are provided in Figure 3.9.

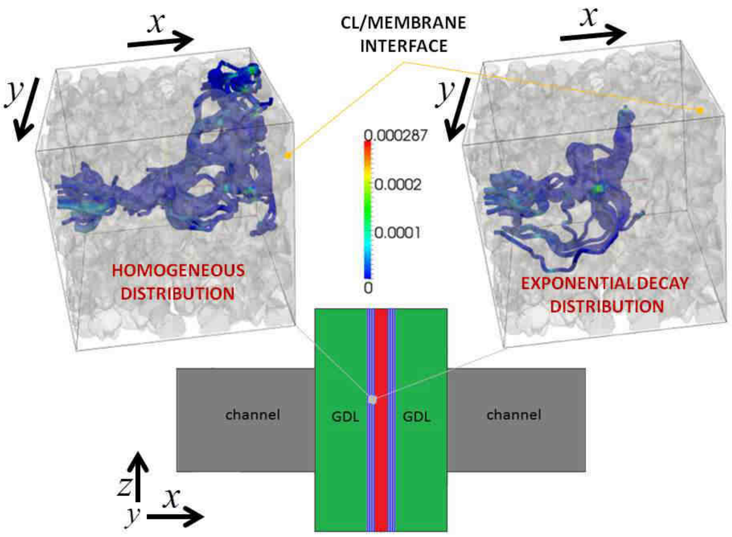


Figure 3.9: A cubic representative configuration of the CL of a HTPEM fuel cell (bottom) is reconstructed by the method proposed in [1]. Fluid flow streamlines through the reconstructed CL porous medium (semi-transparent) corresponding to homogeneous distribution (upper-left) and exponential decay profile (upper-right). Streamline colours indicate the velocity magnitude in dimensionless lattice Boltzmann units.

Changes in terms of both the main flow rate and fluid flow streamlines are observed. In the upper-left part of this figure, when a homogeneous catalyst distribution is adopted in the CL, a considerable number of flow streamlines with non-negligible velocity magnitude can be observed at the CL/membrane interface. On the other hand, much fewer streamlines are able to reach the same interface when an exponential decay distribution of the catalyst is adopted in the CL. In fact, in the upper-right part of Figure 3.9, the flow streamlines for the exponential decay distribution stemming from the same points as in the upper-left plot. As a result, the latter distribution ensures a significant reduction of gas dynamic stresses on phosphoric acid contained within the membrane. Figure 3.10 displays the simulated HTPEMFC overall performance in terms of the polarization curve corresponding to the base case.

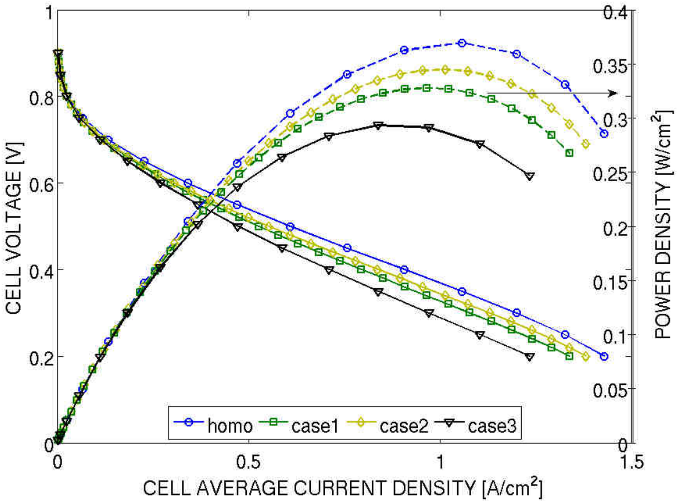


Figure 3.10: Polarization curves resulting from three different configurations of the catalyst particles distribution.

CONCLUSIONS

- A new process has been developed at CNRS to elaborate highly PA doped, cross-linked and reinforced PBI gel membranes, named as generation 2 membrane. The in situ cross-linking reaction and the integration of PBI-based nanofibres into the polymer/PA matrix lead to membranes which are highly conductive, thermally and mechanical stable under the HTPEMFC operating conditions, and they are well adapted to the purpose of ARTEMIS.
- Cathode catalysts FeCo/C, NiCo/C and Pt-Ni/C catalysts were prepared successfully by different conventional chemical methods, and on the basis of ex situ electrochemical characterisation Pt-Ni/C was selected for upscale and electrode development. The anode catalyst Pt/WC-C-CeO_2 shows high tolerance to CO and larger batches were prepared for electrode development.
- A multiscale modelling tool was developed which allows investigation of flow field design details inside catalyst electrode and how different parameters of this modelling tool may be useful to mitigate negative degradation effects during FC operation.

3.3 WP4: MEA ELABORATION AND COMPONENTS SCALE-UP

OBJECTIVES

Work package 4 was dedicated to scale-up of materials from WP3 for integration in MEAs, and development of electrodes and MEAs. It included:

- Membrane scale-up;
- Anode and cathode catalyst scale-up;
- HT-PEMFC electrode development;
- Development of processing for HT-PEMFC MEAs and fabrication of full size MEAs for the ARTEMIS stack.

To achieve deliverables D4.1 in RP1, D4.2-4.3 in RP2 and contribute to MS1 in RP1 and MS3 in RP2.

SUMMARY OF OUTPUT FROM WP4

Several grams of anode and cathode catalyst have been produced and supplied for the development and manufacturing of electrodes and MEAs. Promising performance has been obtained with Pt/WC-C-CeO_2 as anode catalyst and PtNi/MWCNT as cathode catalyst.

The ARTEMIS–2 reinforced and cross-linked membrane preparation process was used to produce homogeneous and defect-free membranes of 400 cm². Several batches of large size membranes for the fabrication of full size MEAs have been produced and delivered.

The composition and the fabrication process of the electrodes, initially made by spraying was changed to a coating process as it was more compatible with the production of the MEA. Successive generations of MEAs comprising all commercial components, commercial electrodes and ARTEMIS membrane, commercial anode, ARTEMIS cathode and ARTEMIS membrane were developed. The performance of the final configuration MEA exceeds the target of 0.5 W/cm² at 1 A/cm² in 25 cm² single cell tests and exceeds the best state-of-the-art. Assembling parameters and sub-gasket were re-worked to fabricate MEAs with the stack design and full size MEAs were supplied for the manufacturing of the final stack.

DETAILED SUMMARY OF ACHIEVEMENTS IN WP4

Membrane Fabrication for MEAs and Stack

Reinforcement material of electrospun and cross-linked PBI nanofibres was prepared using high productivity range electrospinning equipment, allowing the production of 1000 cm² surface web. Polymer was electrospun onto a graphitised polyethylene support to avoid damage during removal from the collector and transfer to the membrane. Membranes were prepared according to final formulation for ARTEMIS – 2 membranes (WP3), and the parameters adjusted in order to reproducibly produce 400 cm^2 membranes. Approximately 20 large size membranes were delivered for full size MEA preparation.

Transfer for continuous membrane preparation was not possible due to incompatibility of the polymer solution and support film on the production line. However this did not impede the supply of membranes for that were continued to be fabricated batch-wise.

Catalyst Elaboration for MEAs and Stack

The preparation process developed in WP3 for anode and cathode catalysts which had led to preparation of approximately 0.4 g of catalyst in a single batch was modified to allow fabrication of 4 g catalyst batches.
Anode catalysts for hydrogen and H_2/CO oxidation were prepared from H_2PtCl_6, using “polyol” methods:

Batch [1]: Pt/Vulcan® XC72R (41 wt% metal), “polyol I”. Pt crystallite size 2.0 ± 0.2 nm.
Batch [2]: Pt/Vulcan® XC72R (53 wt% metal), “polyol”. Pt crystallite size 2.8 ± 0.2 nm.
Batch [3]: Pt/WC-C-CeO_2 (34 wt% metal). Pt crystallite size 2.3 ± 0.5 nm.
Batch [4]: Pt/WC-C-CeO_2 (50 wt% metal). Pt crystallite size 2.8 ± 0.2 nm.
Batch [5]: Pt/WC-C-CeO_2 (50 wt% metal). Pt crystallite size 2.1 ± 0.3 nm.

Cathode catalysts were prepared from H_2PtCl_6 and $NiCl_2$ using an impregnation-reduction method:
Batch [1]: Pt/MWCNTs-c1 (44 wt% metal). Pt crystallite size 2.8 ± 0.2 nm.
Batch [2]: PtNi/MWCNTs-c1 (67 wt% metal, Pt:Ni atomic ratio 2:1). PtNi crystallite size 3.1 ± 0.4 nm.
Batch [3]: PtNi/MWCNTs-c1 (50 wt% metal nominally, Pt:Ni atomic ratio 2:1). PtNi crystallite size 2.2 ± 0.4 nm.

Anode catalysts batches 3, 4, 5 and cathode catalyst batch 2 were delivered for MEA preparation.

Electrode Development

Choice was made to develop Gas Diffusion Electrodes (GDE) by deposting catalyst layers onto a GDL. This choice was motivated by the fact that this approach avoided contact of the acid-doped ARTEMIS membrane with solvents and also avoided excessive compressive stress that leads to the acid desorption. Further, this choice facilitated comparison of the performance of the different improvements of ARTEMIS membranes using a common GDE, and also the performance of the electrodes developed in ARTEMIS with commercial electrodes.

Modifications of the components and parameters of the fabrication processes were introduced into the electrodes and the MEA. The interest of these modifications was evaluated in 25 cm² single cell test fixture.

During RP1, a first generation MEA was developed with an ARTEMIS electrode made with commercial 50 wt% Pt/C catalyst from Tanaka and ARTEMIS membrane. In this first generation MEA, the electrodes were made by spraying the ink onto the GDL. Commercial catalyst was chosen for the fabrication in order to master the process and to understand the influence on the performance of the composition and process parameters. In parallel, work was done in order to develop electrodes with an ARTEMIS catalyst. At the beginning of the RP2, attempts were conducted in order to improve the electrodes made by spraying. However ,in the view of fabricating MEA for the final stack, an ink suitable for coating electrodes, more suitable process in the perspective of producing large amount of material, was developed with Pt/C from Tanaka and the parameters of the coating process (speed, thickness and temperature) were optimised. After several attempts, it was possible to develop electrodes with enhanced performance compared to electrodes made by spraying (same Pt loading, same type and content of binder).

Work was carried out on the composition of electrodes made by coating, with study of the following parameters:

- Type of gas diffusion layer. Performance with SGL 34BC is better than with Freudenberg H2315 C4 for current density lower than 0.3 A/cm² and lower above this value.
- Type of catalyst. PtCo gives lower performance than Pt.
- Content of binder in the electrode. An optimum in the content of PTFE binder was found which significantly improved the performance to close to the best obtained with a commercial (DPS) MEA: slightly higher below 0.4 A/cm² and slightly higher above this value.

Strong improvement of the performance was achieved by optimising the assembling parameters of the components with DPS membranes. Using the optimised ink formulation, self-standing MEAs were then developed using, initially, commercial membranes and then ARTEMIS membranes and ARTEMIS cathode, and integrating an appropriate sub-gasket material and a suitable assembling procedure. Such self-standing MEAs show performance exceeding the best state-of-the-art in 25 cm² cell, Figure 4.1

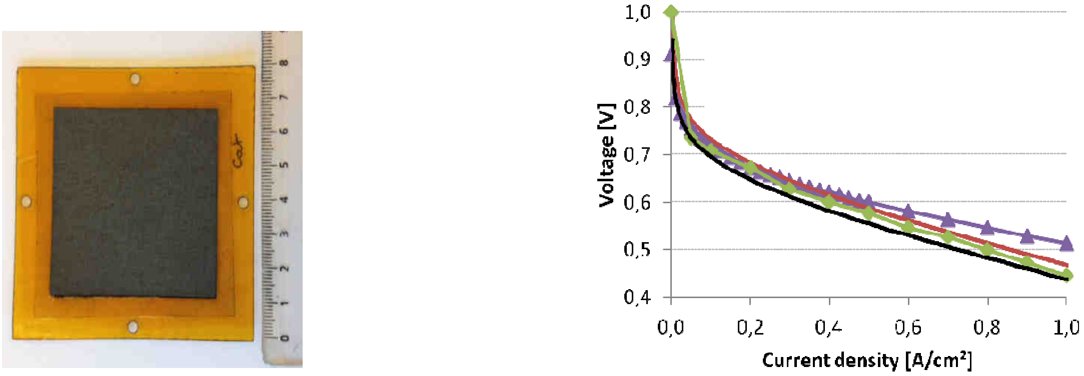


Figure 4.1: (Left) Self-standing ARTEMIS MEA. (Right) Polarisation curves obtained with different MEA. 160 °C, H_2/Air, 1.1/1 bar, no humidification, stoichiometry 1.5/2.5. Black: DPS MEA, green: BASF Celtec P1000, red: ARTEMIS electrodes and DPS membrane, violet: ARTEMIS cathode and membrane, DPS anode.

The composition of this MEA is described in D4.2. Corresponding to the second generation MEA, made with partners' improved and down selected materials, it was selected to be implemented in the final 0.3 kW stack, on the basis of the results obtained during tests in 25 cm² single cell test fixture.

Electrodes were up-scaled and work carried out on the assembling procedure of the real size MEA, particularly on the integration of the ARTEMIS membrane and the sub-gasket, the alignment of the different components and on the assembling parameters. The fabrication of real size MEA without defects required several trials. Such MEAs were supplied for optimisation of assembling pressure of the stack and to check the performance of the first real size MEAs. The really first large size MEAs were fabricated and supplied at M31, Figure 4.2. 8 real size MEAs were supplied from M32 to M38 for preliminary tests. They enabled understanding of the behaviour of the stack and the MEA, and helped to improve the MEA assembling. From M38 to M39, 8 real size MEAs, Figure 4.2, were fabricated for the manufacturing of the final stack. Final MEA production is described in D4.3.

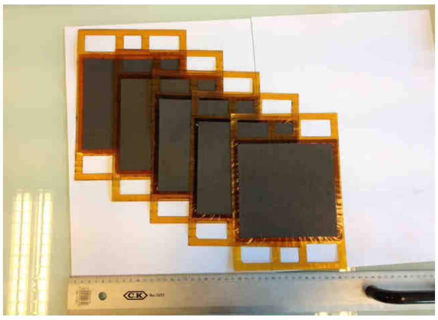


Figure 4.2: Second generation 200 cm² MEAs.

The performance obtained with the second generation MEA in 25 cm² single cell and in the 200 cm² stack, is compared to those obtained with DPS MEA in the same configuration, Figure 4.3. The performance of the ARTEMIS MEA in the stack is only slightly lower than in the single cell. In both configurations and sizes, the second generation ARTEMIS MEA gives higher performance than commercial state-of-the-art DPS.

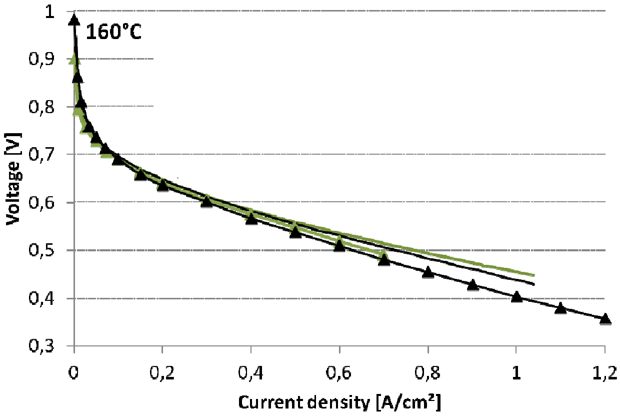


Figure 4.3: Polarisation curves with 25 and 200 cm² DPS MEA and ARTEMIS MEAs. 160 °C, H_2/Air, 1.1/1 bar, no humidification, st. 1.5/2.5. Black: DPS MEAs, Green: ARTEMIS MEAs. Triangles: 200 cm^2, no symbol: 25 cm^2.

Electrodes developed with ARTEMIS anode and cathode catalysts: Anodes containing the ARTEMIS catalysts Pt/WC and Pt/WC-C-CeO_2 (Figure 4.4) were developed and tested. According to the 25 cm^2 fuel cell tests, Pt/WC-C-CeO_2 gives better performance than Pt/WC, although lower than commercial Pt/C in pure hydrogen. The CO tolerance to CO of this catalyst is described in WP6. In addition cathodes developed with ARTEMIS PtNi/MWCNT catalyst show promising performance, Figure 4.4.

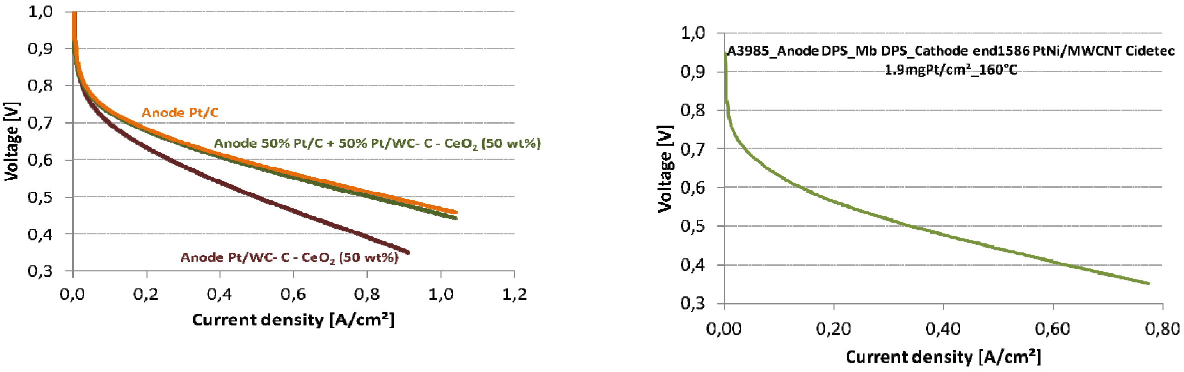


Figure 4.4: Left: Polarisation curves obtained with 25 cm^2 MEA made with anode containing Pt/C catalyst from Tanaka and ARTEMIS Pt/WC- C - CeO_2 catalyst. Right: Cathode containing ARTEMIS PtNi/MWCNT. 160°C, H_2/Air, 1.1/1 bar, no humidification, st. 1.5/2.5.

Pt gradient electrodes: The specifications of these electrodes were defined according to results from the modelling in WP3, where it was concluded that electrodes with a gradient of Pt concentration decreasing from membrane to the GDL would have better durability with only a small decrease in performance (~10%). Electrodes and MEAs were prepared (Figure 4.5) and tested. According to the fuel cell tests, the performance of the electrode with increasing Pt concentration from GDL to membrane is much higher than that of the electrode with decreasing concentration gradient.

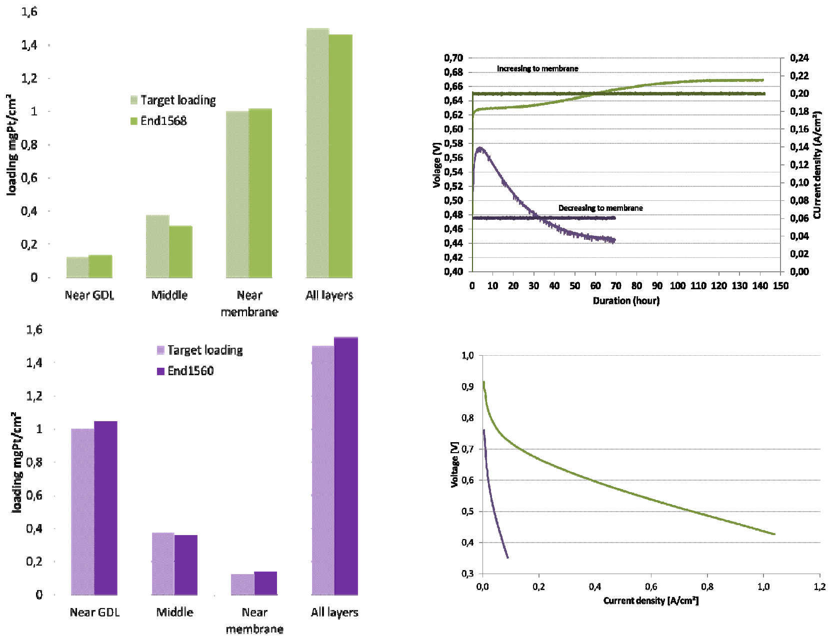


Figure 4.5: Left: Real and targeted Pt loading of each layer constituting electrodes with Pt concentration gradient.
(Top) Increasing and
(Bottom) Decreasing from GDL to membrane.
Right: (Top) Evolution of current density and voltage during break-in; (Bottom) polarisation curves obtained with MEA made with cathode having increasing (green) or decreasing (violet) Pt concentration gradient from GDL to membrane. Membrane and anode from DPS. 160°C, H_2/Air, 1.1/1 bar, no humidification, st. 1.5/2.5.

The discrepancy between what was expected from the model and the experimental result is ascribed to the difference in real and computed distribution of phosphoric acid in the electrodes. In real electrodes, the membrane is the source of phosphoric acid. Consequently, there is a gradient of phosphoric acid increasing from membrane to GDL. The model has been modified to take into account a distribution of phosphoric acid in the electrode as close as possible to the real one. Since good performance can only be achieved if a high surface of Pt can be reached by protons and oxygen at the cathode, this condition is fulfilled for the electrode with Pt concentration gradient increasing from GDL to membrane and not in the reverse case, for which Pt can not be easily reached by protons.

CONCLUSIONS

- Optimisation of solution viscosity and composition, casting and thermal curing parameters have led to a reproducible process for elaboration of large size highly acid doped, cross-linked and reinforced PBI membranes (ARTEMIS – 2 membrane). This process was used to prepare and supply membranes for full size MEA development.
- ARTEMIS anode (Pt/WC-C-CeO_2) and cathode (PtNi/MWCNT) catalysts have been scaled up to a batch size of 4 g. These catalysts have been used in MEAs, with promising results.
- Transfer of membrane fabrication to a small scale production pilot line was initiated using the composition of a first generation, non-reinforced membrane, however it was not finalised since the polymer solution was not compatible with the supports available on the pilot line (D4.4).
- Optimisation of the ink slurry, deposition process, electrode composition and assembling parameters have led to a second generation of ARTEMIS MEAs comprising ARTEMIS membrane and cathode (commercial catalyst) and commercial anode. The composition of this MEA is described in D4.2 and corresponds to milestone MS3. Its performance exceeds the target of 0.5 W/cm^2 at 1 A/cm^2 in 25 cm^2 single cell tests, and the best state-of-the-art and durability exceeds 2000 hours (see WP6).
- MEAs have been developed to investigate predictions from modelling regarding favourable catalyst layer structure.
- Sub-gasket and assembly parameters were investigated in the scale-up from 25 cm^2 to full size (200 cm^2) MEAs. Eight full-size MEAs were supplied for the short stack (D4.3).

3.4 WP5: STACK CONCEPTION, COMPONENTS FABRICATION & VALIDATION

OBJECTIVES

Work package 5 was dedicated to simulation of HT-PEMFC stack operation as a range extender and development of a high temperature PEMFC stack. It included:

- Development of simulation tools;
- Development and selection of stack components including seals, bipolar and end plates;
- Design, construction and test of ARTEMIS stack (0,3 kW, 3 kW).

To achieve deliverables D5.1 in RP1, D5.2-5.3 in RP2 and achieve MS3 in RP2.

SUMMARY OF OUTPUT FROM WP5

In the simulation task, the goal was to simulate stack operation under automotive range extender conditions, based on FC system component models from Artemis partners. Vehicle models were developed in order to assess the effects of the introduction of an HT-PEMFC on the vehicle range through the simulation of different driving cycles. Considering a fuel cell stack of 3 kW, two different solutions were analysed 1) On board generation for light commercial vehicles, where the fuel cell is used to supply the average power due to the auxiliaries loads; 2) Charge sustaining suitable for passenger cars, where the fuel cell is used to supply the average power for the traction. For the first application, the vehicle runtime was improved by about 11% for the NEDC, and by about 19% for the Asterics driving cycle, with respect to the full electric vehicle. For the second application, a 3 kW fuel cell was shown to be able to ensure the battery charge sustaining in the cases of UDC and Artemis Urban driving cycles.

Composite bipolar plate and sealing materials were developed that can operate at demanding high temperature conditions. Properties of bipolar plates made with mass-production technology have been tested on resistance, temperature stability, corrosion, leakage and acidity resistance to check compatibility. A second cell plate formulation was processed and tested for performance. Mechanical tests were performed to confirm the improved mechanical properties of this material. Seal material was chosen and the required seal thickness was calculated. The hardware components design was completed.

After preliminary performance tests on single cells to optimise the HT-PEM MEA production process, stack assembly and the HT PEMFC test station, a 4-cell 300 W stack was assembled and tested. It was confirmed that this stack produces 0.3 kWe at 160 °C for currents over 165 A (825 mA/cm^2) and at 180 °C for currents over 140 A (700 mA/cm^2).

Using the data from the 4-cell 300 W stack, a 3 kW HT-PEM stack was configured. It was determined that 48 cells of average performance are required to produce 3 kWe at 160 °C in the range of 125-160 A (BoL to EoL) and at 180 °C in the range of 110 A (BoL) to 140 A (EoL). Based on 15% voltage decay as end-of-life condition this configuration allows for the output of 3 kWe from BoL to EoL in a suitable current range. These metrics are consistent with the application requirements.

DETAILED SUMMARY OF ACHIEVEMENTS IN WP5

Simulation tools and their application to stack operation under range extender application

Vehicle models were developed with the simulation software GT-Suite to assess the effects of the introduction of an HT-PEMFC on the vehicle range through the simulation of different driving cycles. Considering a fuel cell stack of 3 kW, two different solutions were analysed:

- On-board generation for light commercial vehicles (e.g. FIAT Iveco Daily 35S), where the fuel cell is used to supply the average power due to the auxiliaries loads;
- Charge sustaining suitable for passenger cars (BMW i3), where the fuel cell is used to supply the average power for the traction.

On-board generation in an electrical automotive application: The modelling of the vehicle was performed through a quasi-static approach: a driver model (PID controller) compares the target vehicle speed with the actual speed and generates a power demand profile in order to follow the target vehicle speed profile, by solving the longitudinal vehicle dynamics equation. However, although system dynamics was taken into account, the behaviour of the main devices (fuel cell, electric motor-generator, batteries) was described using steady state performance maps. The external load on the vehicle was calculated by means of the main vehicle characteristics and of the vehicle resistance drag measured through the coast down test. Following validation of the model, the effects of the introduction of a Fuel Cell-Range Extender (FC-RE) on the vehicle range were studied. The FC-RE supplies constant power (1.3 kW) equal to the electrical loads of the vehicle (pumps, radiator, fan, lights, cabin heater, etc.) and the fuel cell accessory loads (coolant pump and air compressor), during drive and non-drive time, so reducing exposure to load transients.The battery provides traction power and satisfies load transients.

A thermal model of the fuel cell was developed in order to capture the fuel cell behaviour during the warm-up phase. The benefits of the introduction of a FC-RE on the FIAT Iveco Daily 35S were investigated through numerical simulation. Figure 5.1 shows the model results over NEDC and Asterics driving cycles, indicating that when the FC-RE provides 1.3 kW of electrical power to supply the accessory loads, the vehicle runtime is improved of about 11%, for the NEDC (19% for the Asterics) respect to the full electric vehicle. The simulations were performed with a hydrogen tank capacity of 30 litres, with a maximum pressure of 350 bar.

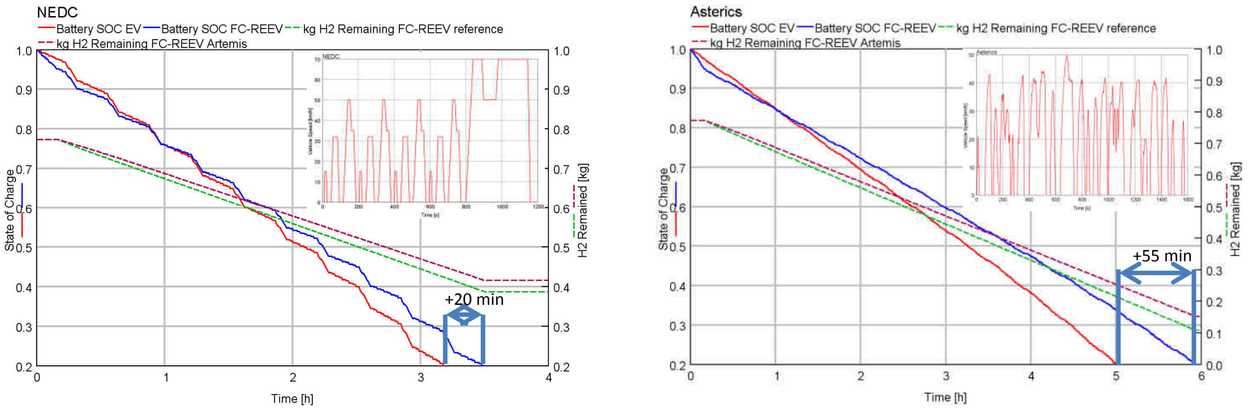


Figure 5.1: Results of simulations over NEDC and Asterics driving cycles for a FIAT Iveco Daily 35S: on-board generation.

A sensitivity analysis was carried out to assess the impact on the hydrogen consumption of the increased performance obtained with the final ARTEMIS MEA. This analysis showed a 5% decrease in hydrogen consumption passing from the baseline fuel cell to the advanced ARTEMIS type, thanks to its higher conversion efficiency.

Range extender: The BMW i3 was chosen as a study case for the charge sustaining application. In this configuration, the fuel cell provides power to the battery maintaining a constant State of Charge. The fuel cell charge sustaining capabilities on different urban driving cycles were analysed through the numerical simulations. A 3 kW fuel cell is able to ensure the battery charge sustaining only in the cases of UDC and Artemis Urban driving cycles; conversely, in the case of WLTC City Cycle, the fuel cell is not able to guarantee a perfect charge sustaining and only 6 cycles are ensured before the battery reaches the lower allowed SOC level (equal to 0.2). Table 5.1 shows the performances of the Artemis fuel cells (reference, ARTEMIS–1 (intermediate) and ARTEMIS-2 (advanced)) on the Urban Driving Cycle (UDC) of NEDC. The range extension caused by the use of a fuel cell depends on the hydrogen tank volume and pressure and on the fuel cell characteristics. For example, using the reference fuel cell with a hydrogen tank of 30 L at 350 bar, the range increase is of 90 km on the UDC, while an increase of 102 km is found using the ARTEMIS advanced fuel cell. In fact, the advanced fuel cell is characterised by greater efficiency and lower hydrogen consumption, about 13% less than that of the reference fuel cell.

Table 5.1: Comparison between ARTEMIS FC performances on NEDC – UDC driving cycle

	FC Baseline	**FC Intermediate**	**FC Advanced**
Trip distance [km]	4.07	4.07	4.07
H_2 consumption [g/Cycle]	32.48	30.06	28.74
Specific H_2 consumption [g/km]	7.98	7.39	7.06
Fuel tank capacity [g]	724	724	724
Fuel tank equivalent cycles	22	24	25
CS mode allowable cycles	unlimited	unlimited	unlimited
Final cycles	22	24	25
Battery range [km]	150	150	150
H_2 range [km]	90	98	102
Final SOC	0.2588	0.2596	0.26
Remaining H_2 [g]	10	3	6
Total Range [km]	240	248	252

The fuel cells current density, voltage, efficiency and hydrogen consumption along the UDC are presented in Figure 5.2, where it is seen that the advanced fuel cell works at lower current density and higher voltage than the baseline and intermediate ones. This reduction in current density decreases the hydrogen consumption making the advanced fuel cell work at a higher efficiency.

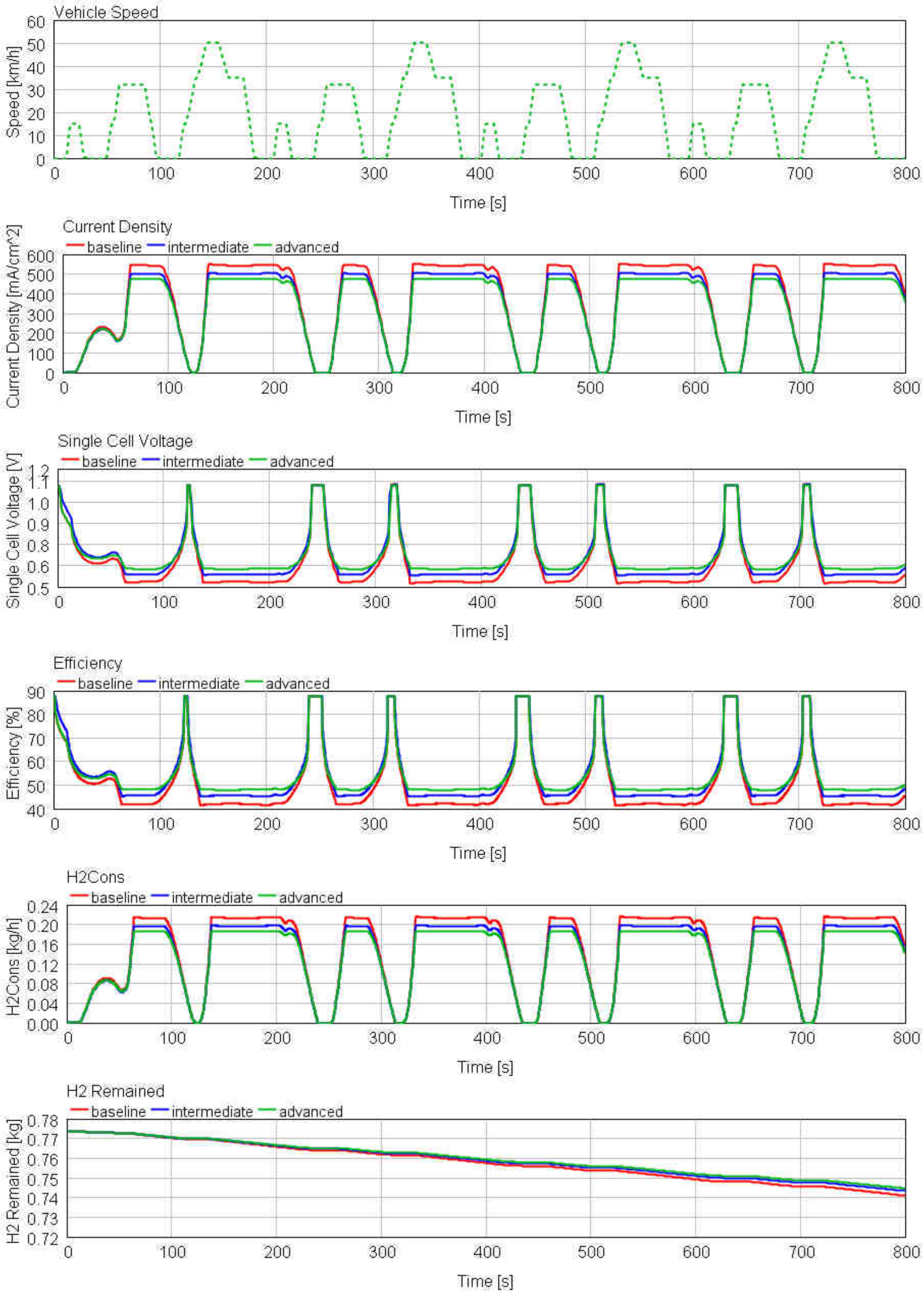


Figure 5.2: Comparison between ARTEMIS FC performances on NEDC – UDC driving cycle.

Stack components

ARTEMIS cell plates are carbon/polymer composites. Accelerated stress tests were developed and applied in RP1 to HT-1 plates to assess their suitability for HT PEMFC, and the results were used to modify the specific composition of cell plates to achieve an improved electrical conductivity, and a higher process temperature was used, providing plates HT-2. Mechanical tests were performed on dumbbell shapes prepared from flat cell plate sheet. Elongation versus stress and flexural strain are shown in Figure 5.3, at room temperature and at 70, 180, 200 and 230 °C. Mechanical properties for bending and tension fulfil requirements.

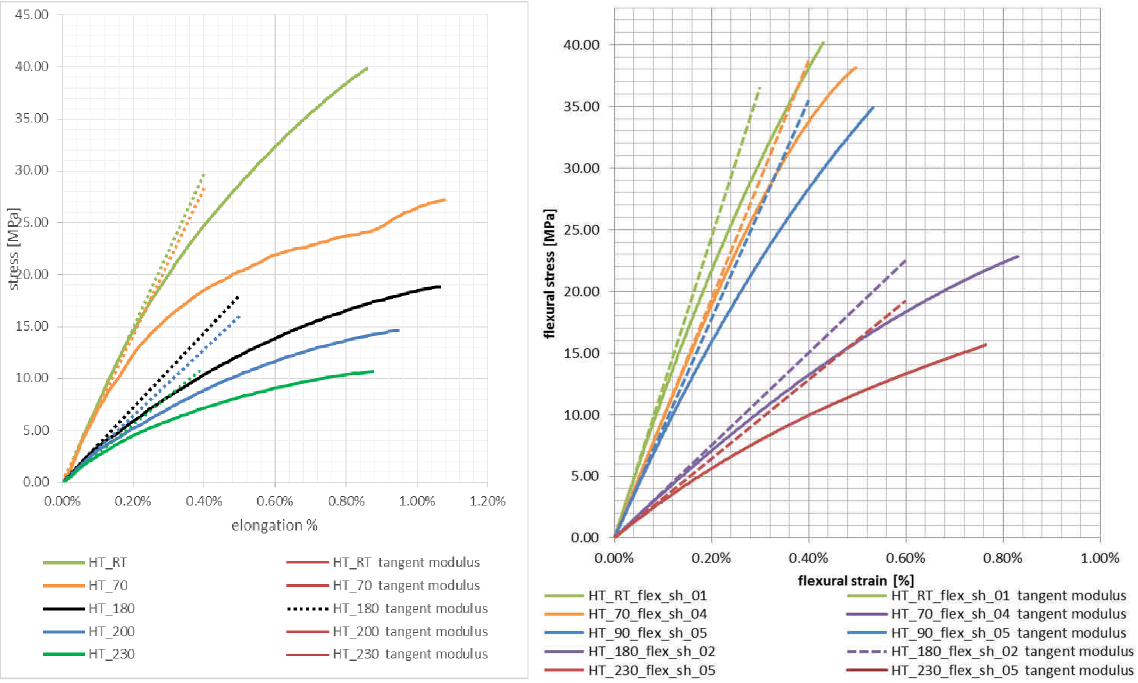


Figure 5.3: Left: tensile properties, right: flexural properties of ARTEMIS HT-2 cell plates

HT cell plates were produced from HT-2 for the 0.3 kW stack. Specific materials were selected for seals and other stack hardware. Seal compression was tested and found to meet requirements, although leak rates were found to be slightly higher then expected. A second batch of seals with increased thickness was used.

300 W Stack Assembly and Validation

HT-PEM stacks were designed on the basis of MEA performance observed in WP4 and the typical operating conditions defined in WP2. The component design and materials used had to be suitable for large thermal cycles, resulting in stringent materials specifications, Figure 5.4. The stack was assembled several times with di to 4 cells, compression was optimised and the HT plates were validated.

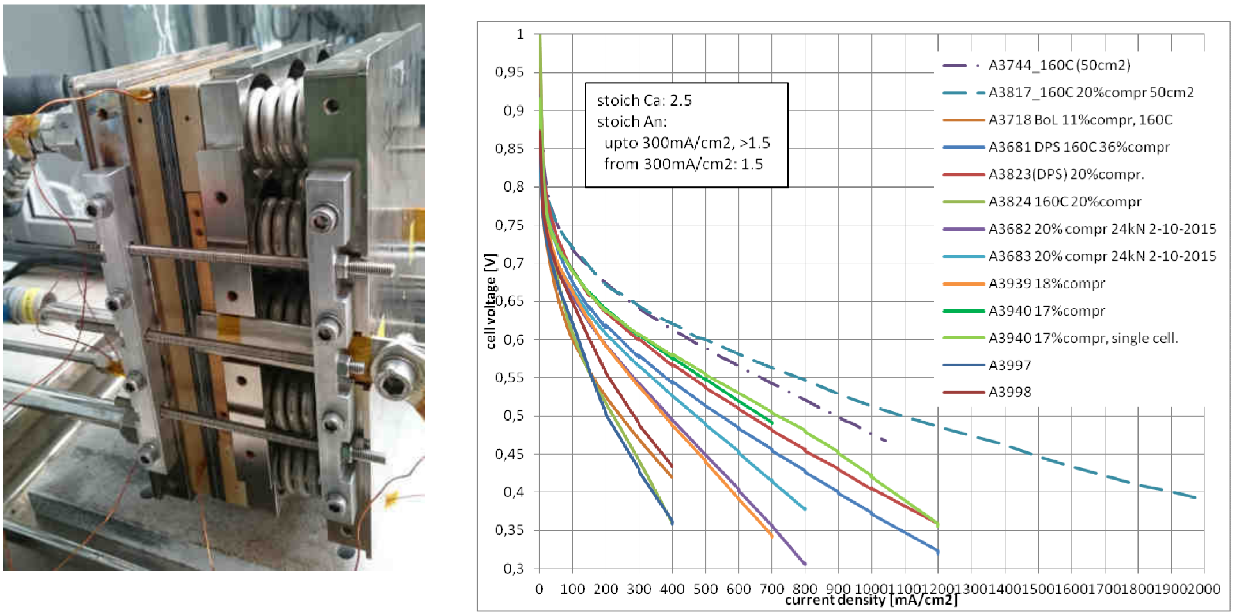


Figure 5.4: (left): HT-PEM 200 cm^2 platform designed and constructed in ARTEMIS; (right) full size single cell performance tests on reference and ARTEMIS MEAs recorded in stack hardware.

Following this initial calibration and optimisation, a 0.3 kWe prototype stack was constructed using the stack components developed in Task 5.2 (reported in Deliverables D5.1 and D5.2) and MEAs developed in WP4. Based on the results from this 0.3 kW stack, a 3 kWe prototype stack was configured as reported in Deliverable D6.5.

The polarisation curves shown in Figure 5.4 were recorded after >100 h conditioning at 160 °C. The project target was to produce MEAs with a performance at least as good as the best known commercially available MEA, shown by the bright red curve in Figure 5.4 (A3823 DPS), which is an MEA which fully comprises DPS components, assembled following the same procedure as the ARTEMIS MEAs and tested in the same 200 cm^2 hardware under the same conditions. The 50 cm^2 MEAs (3744 and 3817) have significantly higher performance than the 200 cm^2 DPS MEA. The 200 cm^2 MEAs have significantly lower performance than the 50 cm^2 variants, however MEA A3940 has a performance above the set target. MEAs with the same components can result in significantly different performance. The reason for this is unknown; however, the probable cause was identified in the MEA assembly process, with further optimisation required for the full size cell assemblies.

A 4-cell stack was assembled with MEAs A3940, A3962, A3964 and A3972 and was tested at 160 and 180 °C, Figure 5.5. IV curves are shown for 160 °C only, since at 180 °C they follow the same trend, while the stack power output is shown for 160 and 180 °C. This stack produces 300 W at 160°C for currents over 165 A (825 mA/cm^2) and at 180 °C for currents over 140 A (700 mA/cm^2). The average cell voltage from the 300 W stack at 160 °C was used to calculate the average cell power, and the number of cells required for a stack with 3 kWe power output in a suitable current range was calculated to be 48.

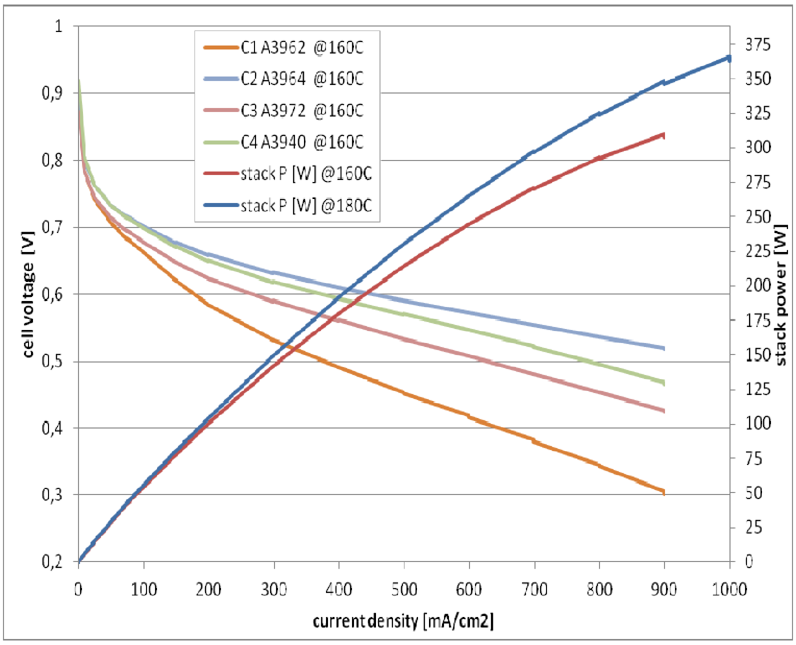


Figure 5.5:

Results of performance tests on 4-cell 0.3 kWe stack at 160 °C, and stack power at 160 and 180 °C.

A performance decay of 15% cell voltage was applied to the beginning of life data as an end-of-life condition and the power output for a 48 cell stack at end of life was calculated. Figure 5.6 shows that when operating at 160 ℃ increase in current from 125 A at BoL to 160 A at EoL would be required in order to produce 3 kWe (from 110 to 140 A at 180 ℃. Extrapolation of experimental voltage decay results occurring after 1 000 h and 2 000 h single cell operation with the ARTEMIS range extender protocol on 50 and 25 cm^2 cells with ARTEMIS MEAs suggests that such 15% voltage loss would occur after some 11 250 h operation.

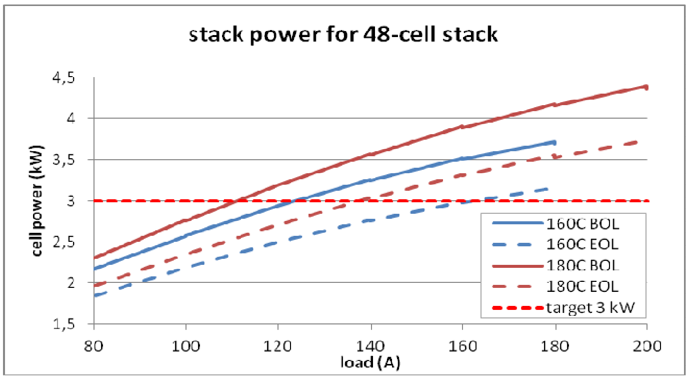


Figure 5.6: Calculated stack power output at BoL and EoL (15% decay) at 160 and 180 °C in applicable load range. 3 kW target is indicated with red dotted line.

CONCLUSIONS

- With a 3 kW HT-PEMFC in a light commercial vehicel, simulation shows that vehicle runtime improves by 11 and 19% for the NEDC and Asterics driving cycles respectively, with respect to the full electric vehicle.
- A 3 kW HT-PEMFC in a passenger car ensures the battery charge sustaining in the cases of UDC and Artemis Urban driving cycles
- Cell plates developed in ARTEMIS have been validated for HT-PEMFC
- Full size ARTEMIS MEAs have improved performance over the state of the art
- A 4-cell stack with ARTEMIS MEAs produces 0.3 kWe at 160 °C for currents over 165 A (825 mA/cm^2) and at 180 °C for currents over 140 A (700 mA/cm^2) and the results used to configure a 3 kW stack.
- Full size ARTEMIS MEAs have lower performance than 50 cm2 MEAs and further optimisation of assembling is needed to reach their full potential.

3.5 WP6: FUEL CELL TESTING: SINGLE CELLS AND STACK

Objectives

MEAs are characterised at single cell and stack levels in Work package 6, both for their performance and operation under range extender protocol. It included:

- Single cell testing of MEAs delivered from WP4;
- Stack testing of the stack delivered from WP5;
- Perform cell and stack testing using WP2 protocols: IV curves and simulated range extender operation.

To achieve deliverables D6.1-6.2 in RP1, D6.3-6.5 in RP2.

Summary of Output from WP6

Numerous experiments have been conducted to validate the developed materials and the process feasibility at the single cell and the stack level. MEAs prepared using the ARTEMIS – 2 membrane and ARTEMIS cathode have been evaluated at 25 and 50 cm^2 level. Performances are higher than those of commercially available MEAs, with notably high voltage at high current density (25 % higher at 1.2 A/cm^2). A 1000 hours evaluation has been conducted using the range extender drive cycle on a MEA with the ARTEMIS-2 membrane and commercial electrodes, giving -8 µV/h voltage decay. Under hold current density condition (0.2 A/cm^2) an ARTEMIS–2 MEA underwent 8 µV/h loss after 1000 h. CO tolerance of the HT PEMFC single cells was investigated, and improved CO tolerance of an MEA with an ARTEMIS anode catalyst has been shown to hold promise for a future generation of ARTEMIS MEAs.

The 4-cell stack comprising ARTEMIS–2 MEAs was tested and was submitted to the range extender protocol.

Detailed Summary of Achievements in WP6

Single cell testing

The performance of ARTEMIS MEAs have been evaluated under continuous operation and on load cycling. Space does not allow a comprehensive presentation of the results obtained, and the report is limited here to providing some examples. Figure 6.1 provides results obtained in RP1 using ARTEMIS–1 membranes and commercial (DPS) electrodes, compared with commercial MEAs.

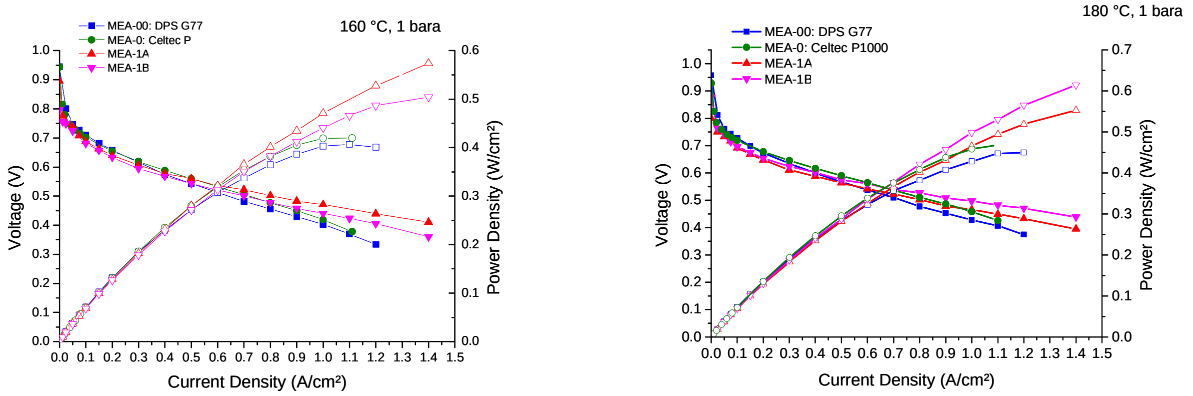


Figure 6.1: I/V characteristics of ARTEMIS-1 membrane based MEAs, comparison with reference MEAs at 160 and 180 °C. 160 °C H$_2$/air at 1 bara (anode and cathode), stoichiometries of 1.5/2.5. MEAs 1A and 1B comprise 80 µm membranes, significantly thinner than those of MEAs 0 and 00.

The ARTEMIS–2 MEA comprises the ARTEMIS-2 membrane, ARTEMIS cathode (commercial catalyst) and commercial anode, with optimised MEA assembly conditions. I/V characteristics of ARTEMIS-2 MEA and reference MEA-00 (commercial DPS G77 MEA) are compared in Figure 6.2. In the regions of ohmic and concentration polarisation, the measured voltage of ARTEMIS–2 MEA is higher over the whole range of current densities. This large voltage gain is likely due to the use of a thinner membrane, a better acid retention and a better membrane-electrodes interface for the ARTEMIS MEA.

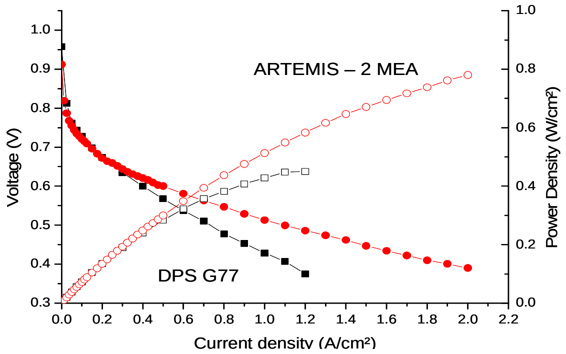


Figure 6.2: I/V characteristics of ARTEMIS–2 MEA compared to DPS reference MEA.

Continuous operation shows little voltage decay, despite two test stand stop/start events at 523 and 670 h, Figure 6.3, Figure 6.4.

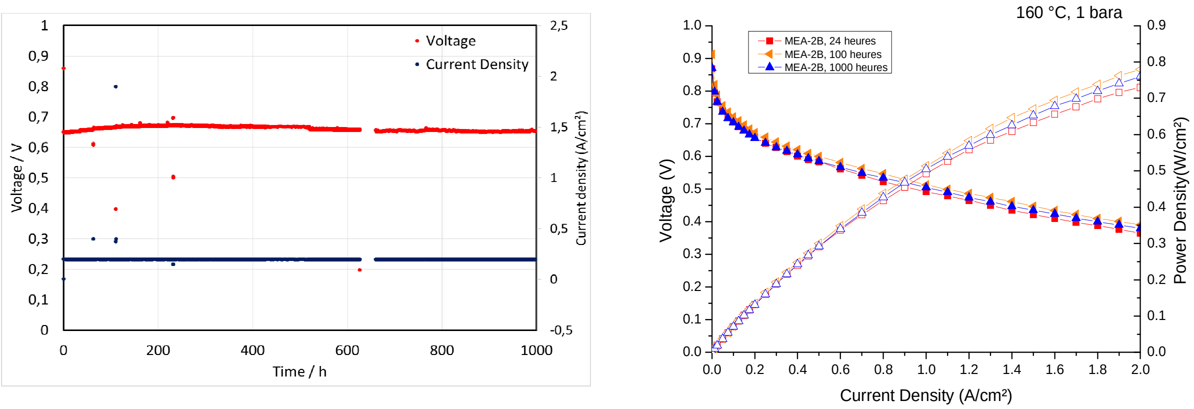


Figure 6.3: (left) ARTEMIS–2 MEA continuous operation at 0.2 A/cm² (right) I/V curves after 24, 100, 1000 h.

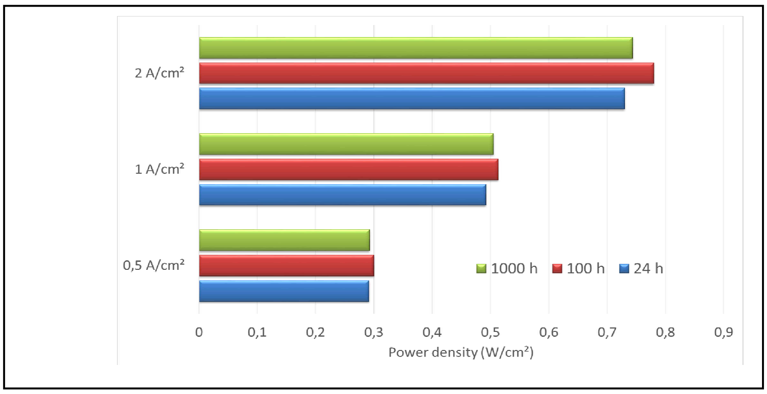


Figure 6.4: Power densities of ARTEMIS-2 MEA after continuous operation for 24, 100, 1000 h at 160 °C. The power densities at 0.5, 1 and 2 A/cm² are, respectively, centred on 0.3, 0.5 and 0.75 w/cm², regardless of the test duration, consistent with the ca. 7 µV/h voltage decay.

Durability tests were conducted on MEA-2A comprising the reinforced ARTEMIS–2 membrane by starting cycling (35 min at 0.2 A/cm², 25 min at 0.4 A/cm² and 5 min at 0.6 A/cm²) after 270 h continuous operation at 0.2 A/cm² for a total operation time of 1000 h, Figure 6.5. From the beginning of the cycling test to the end of the test, the average voltage decay over the cycling period (from 270 h to 1000 h) is ca. 8 µV/h.

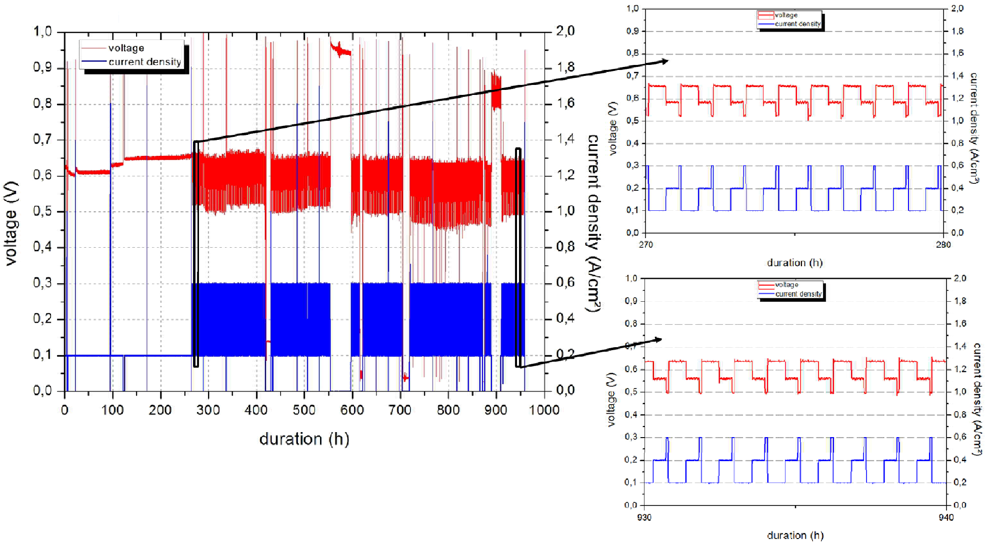


Figure 6.5: I/V characteristics of ARTEMIS-2 membrane based MEA (commercial electrodes) under range extender drive cycle.

Similarly, 2240 h of operation including 2000 h of cycling with ARTEMIS range extender protocol have been demonstrated with an ARTEMIS–2 MEA. The loss in performance was only 3% at 0.2 A/cm² over the 2000 hours of cycling including the effects of start/stop which has huge impact, Figure 6.6. This demonstrates the robustness of the ARTEMIS MEAs. It is important to note that prior to the durability tests, the MEA was intensively characterised in different operating conditions, which explain why the initial voltage at the beginning of the cycling (after 240 hours) is only 651 mV whereas it is around 680 mV after break-in.

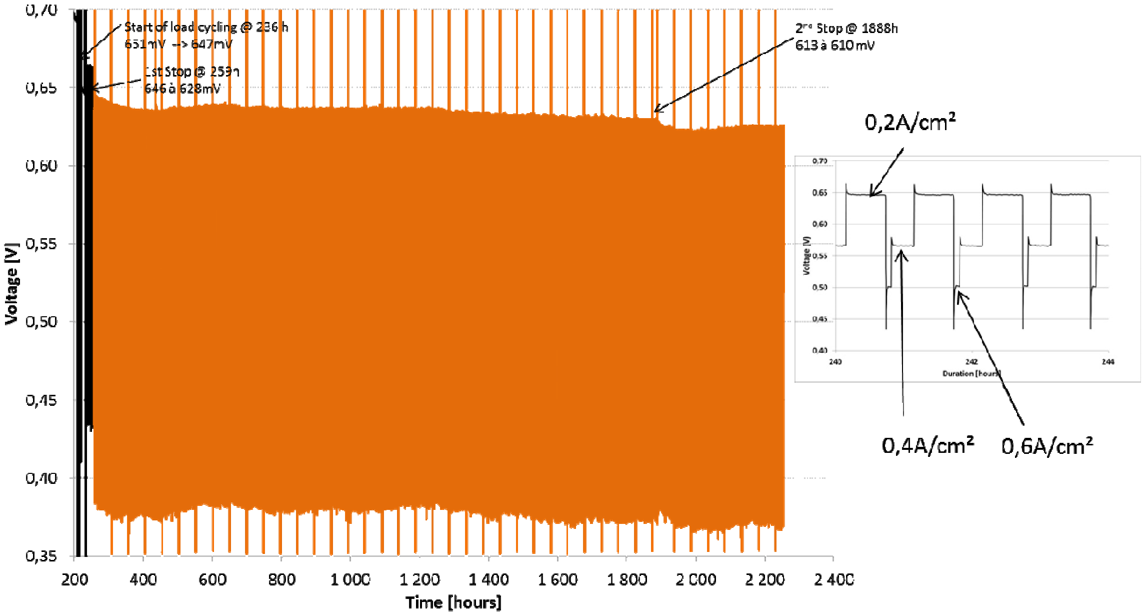


Figure 6.6: Evolution of voltage during cycling durability test with ARTEMIS protocol. (Left) Cell voltage over >2000 hours. Note that it is not possible at this scale to show the values of voltage. (Right) Zoom at the beginning of the test for 4 cycles. ARTEMIS MEA comprising ARTEMIS electrodes and DPS membrane. 160 °C, H_2/Air, 1.1/1 bar, non humidification, stoichiometry 1.5/2.5.

It is well-known that the CO tolerance of PEM fuel cells increases with the temperature of operation, and that this is one advantage of HT-PEMFC compared to low temperature technology. The performance obtained with ARTEMIS electrodes (commercial catalysts) is similar to those recorded with DPS electrodes on pure H_2, however an ARTEMIS anode with commercial Pt/C catalyst is more sensitive to CO than DPS electrode, Figure 6.7, showing that improvement in the formulation of the anode catalyst is required. Using the ARTEMIS anode catalyst (Pt/WC-C-CeO_2) in a blend with Pt/C provides improvement of the CO tolerance compared to a DPS electrode, Figure 6.8.

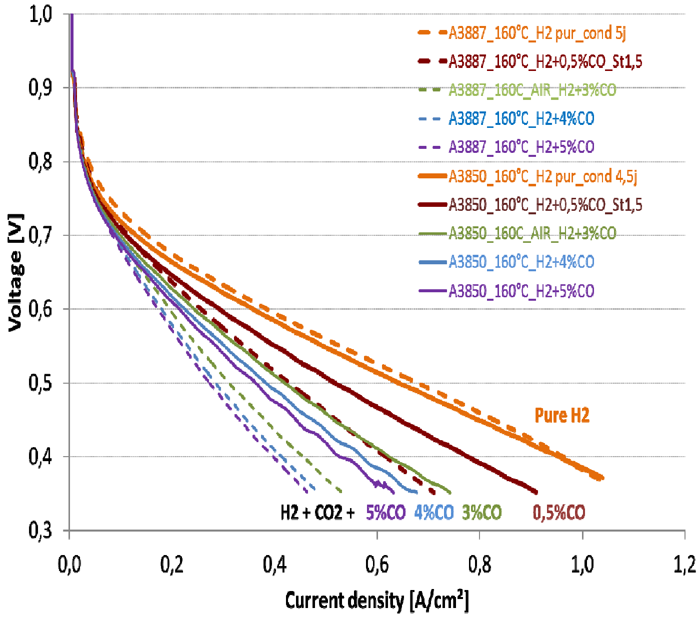


Figure 6.7: Polarisation curves obtained for different CO content in synthetic reformate (21% CO_2 + x% CO + (79 – x)% H_2). A3850: MEA made with DPS components. A38887: MEA with DPS membrane and ARTEMIS electrodes. 160°C, Synthetic reformate/Air, 1.1/1 bar, no humidification, stoichiometry H_2/air 1.5/2.5.

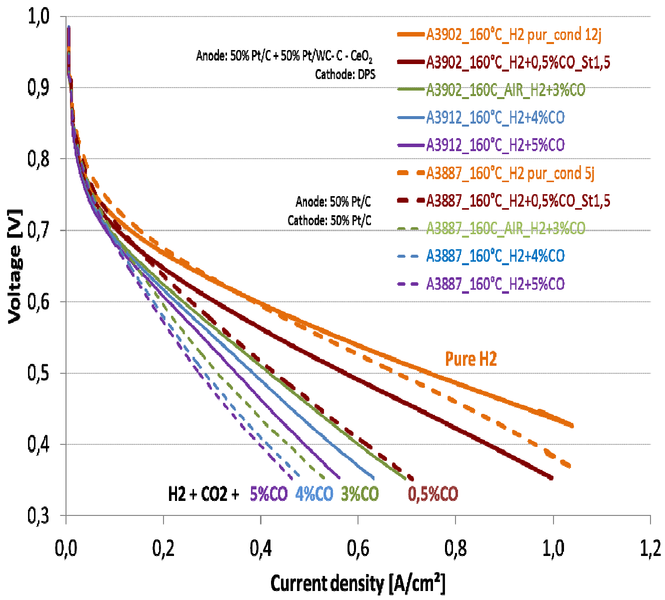


Figure 6.8: Polarisation curves obtained for different CO content in the synthetic reformate (21% CO_2 + x% CO + (79 – x)% H_2). A3887: MEA with DPS membrane and ARTEMIS electrodes (commercial Pt/C). A3902: MEA with DPS membrane and ARTEMIS electrodes, Anode with commercial Pt/C and ARTEMIS Pt/WC-C-CeO_2, Cathode with commercial Pt/C. 160°C, Synthetic reformate/Air, 1.1/1 bar, no humidification, stoichiometry H_2/Air 1.5/2.5.

The durability of the electrodes with a gradient of concentration of Pt was also been evaluated, Figure 6.9, to compare the effect on the durability of the direction of the Pt concentration gradient within the electrode. The composition of these electrodes is described in WP2, where modelling predict higher durability for a decreasing Pt concentration gradient from GDL to the membrane. The MEA has a lower performance than the one with a reverse gradient. The cell voltage, evaluated in galvanostatic mode at 60 mA/cm², gently increases, which can be explained by the leaching of phosphoric acid from the membrane to the electrodes where it can reach area with higher Pt loading. In contrast, with a Pt concentration gradient increasing from the GDL to the membrane decreases during the durability test. Phosphoric acid which is leached out goes in area of the electrodes where the Pt concentration is lower. In this latter case, the durability protocol is the cycling protocol defined in ARTEMIS. It is concluded that an electrode with a Pt concentration gradient increasing from the GDL to the membrane is less durable than an electrode with a Pt concentration gradient in the opposite direction.

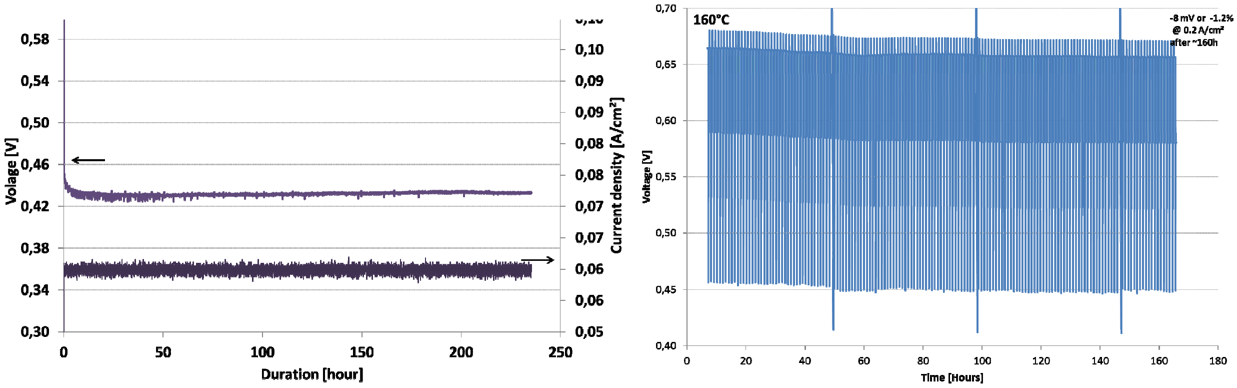


Figure 6.9: Evolution of voltage during durability test. (Left) ARTEMIS cathode with decreasing Pt concentration gradient from the GDL to the membrane and constant load at 60 mA/cm². (Right) ARTEMIS cathode with increasing Pt concentration gradient from the GDL to the membrane and ARTEMIS cycling durability protocol. DPS Membrane and anode. 160°C, H_2/Air, 1.1/1 bar, no humidification, st. 1.5/2.5.

Stack testing

The initial stack testing was interrupted by a leak of cooling fluid that penetrated into one of the cells and contaminated it. This cell had to be removed, and it was replaced by another, lower performing cell before the stack was submitted to further testing. While the original 4-cell stack (MEAs c1 A3940, c2 A3682, c3 A3964 and c4 A3962) had provided the target 309 W at 900 A/cm2 at 160 °C, the reconstructed 4-cell stack (c1 A3972, c2 A3682, c3 A3964 and c4 A3962) provided a maximum power density of 282 W.

The stack durability has been evaluated on application of a simplified range extender protocol. The first test consisted in the cycling of the current density at 0.2 A/cm² for 35 min and at 0.4 A/cm² for 25 min. The evolution of both the stack and cell voltages were recorded, Figure 6.10.

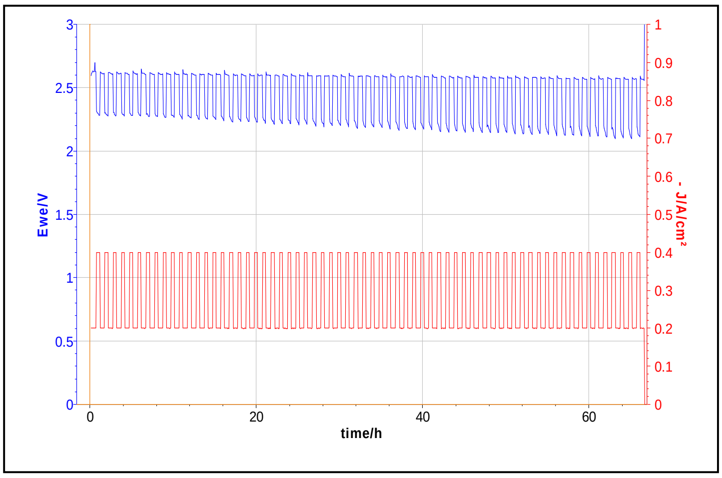


Figure 6.10: Recorded voltages of the ARTEMIS stack under current cycling conditions, 160°C, H_2/Air, 1.1/1 bar, no humidification, stoichiometry 1.5/2.5.

The voltage decays of the stack and the 4 different cells over a period of 70 hours were higher than observed in any of the single cell durability tests, 0.1-0.2 mV/h at 0.2 A/cm^2, and 0.2- 1 mV/h at 0.4 A/cm². The ratio between the values of voltage decay at respectively 0.2 and 0.4 A/cm² is in the range 2.5 – 4 for both the stack and the cells 1, 3 and 4, while this ratio is 7 for the cell 2. A second durability study was conducted on the stack using the range extender protocol defined in ARTEMIS, and the stack and the cell voltages are shown in

Figure 6.11. A leak from cell 4 caused shutdown of the stack, this failure attributed to a pinhole in the membrane. Cell#4 was positioned on the side of the gas inlet.

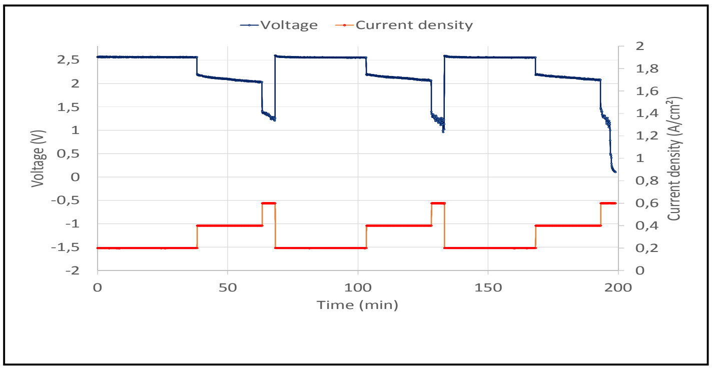
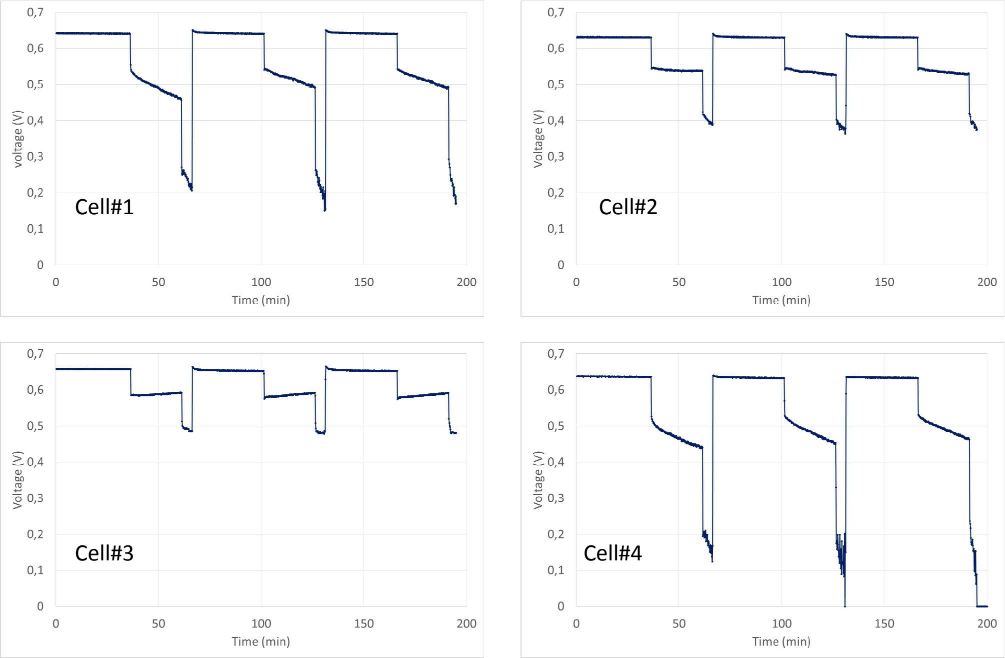


Figure 6.11: Recorded voltage of the stack and the 4 cells under the Range Extender Protocol, 160°C, H_2/Air, 1.1/1 bar, 0%RH, st. 1.5/2.5.



Unfortunately there was not sufficient time in ARTEMIS to completely optimise handling and assembly of large size membranes, MEAs and cells the prototype high temperature stack. However it is important to note that the targets were reached in terms of stack power with only 4 cells, and that the performance in such full size cells exceeds that of commercial MEAs. It is recalled that in smaller (50 cm^2) cells, the performance is clearly significantly higher than commercial MEAs, in particular at high current densities, with very high durability during continuous operation and range extender cycling longevity tests.

CONCLUSIONS

- MEAs from WP4 comprising ARTEMIS materials (membrane and cathode) have single cell (25, 50 cm2) performance largely exceeding commercial MEAs;
- Continuous operation and range extender protocol load cycling have been successfully conducted up to end of test (not end of life), up to 2200 h, with a voltage decay of -8 µV/h;
- The ARTEMIS anode catalyst in a blend ink with a commercial Pt/C catalyst, provides improved CO tolerance in synthetic reformate
- The ARTEMIS 4-cell stack delivers 300 W, however the lifetime was short due to a leak from one of the cells, and improvements are required to improve durability of the stack and full size MEAs.

4. MAIN DISSEMINATION ACTIVITIES, POTENTIAL IMPACT AND EXPLOITATION OF RESULTS

4.1 MAIN DISSEMINATION ACTIVITIES DURING THE PROJECT LIFETIME

Dissemination channels have included a dedicated project website, presentations at national and international conferences, and publications in international scientific journals.

DEDICATED PROJECT WEBSITE

The dedicated project website was one of the main dissemination channels towards the public and the broad scientific community. The website www.artemis-htpem.eu was released at month 6 and has been fully operational since 15th March 2013.

The project website also includes a link to a confidential area where access is restricted to authorised users.

The public section features:

- General description of the project and its objectives
- Information about the consortium and links to partners' websites
- Public documents, such as public deliverables, abstracts of publications, project posters
- Contact information

The web site will be kept open and updated with information on publications and other project output until a year after the project end date, when the situation will be reassessed with the ARTEMIS partners.

ARTEMIS DISSEMINATION EVENT

A highlight of dissemination activities in RP2 was the organisation of a dissemination event/workshop on "Renewable energy for transport application: Automotive pemfc Range extender with high TEMperature Improved meas and Stacks (ARTEMIS)", 30th October 2015, Tashkent, Uzbekistan. This event was led by partner Politecnico di Torino, the leaders of WP7 "Dissemination", with special aim to disseminate the results of the project to the industrial partners (potential end-user such as General Motors Uzbekistan, UzAutoSanoat, General Motors PowerTrain Uzbekistan) as well as public/academic auditorium (researchers, post-doc and PhD students of Turin Polytechnic University in Tashkent, National University of Uzbekistan, Institute of Solar Energy of Academy of Science of Uzbekistan, Tashkent Automobile and Road Construction Institute). The meeting aimed at isolation and discussion of some less-explored problems of development and optimisation of the alternative materials for a new generation of high temperature polymeric electrolyte membrane fuel cell (HT-PEMFC) for transport application. Special purpose was ARTEMIS project results dissemination activity towards to the scientific and public communities. This has been done by presenting invited and short talks for the broad auditorium of specialists from different fields, like research and development scientists and engineers, system producers, hydrocarbon fuels infrastructure developers and final user automotive sector, as well as young researchers working on different areas of modern material science, physics and chemistry.

ARTEMIS dissemination at the Workshop:

- **Overview of ARTEMIS: Automotive pemfc Range extender with high TEMperature Improved meas and Stacks**, Pietro Asinari, Politecnico di Torino
- **Improved PEM fuel cell range extender testing in pure electrical vehicles**, Sabino Caputo, Politecnico di Torino and Centro Ricerca FIAT

- **Multiscale modelling to boost durability and performance of high temperature fuel cells for automotive application**, Uktam Salomov, Politecnico di Torino and Turin Polytecnic University in Tashkent
- **Novel materials for a HT-PEMFC stack for operation as automotive range extender**, Y. Nedellec, A. Kreisz, N. Donzel, J. Rozière, D. J. Jones, A. Morin, C. Nayoze, F. Alcaide Monterrubio, M. Smit, P. Asinari, U. Salomov, A. Piu
- **Performance of HT-PEMFC on operation as automotive range extender**, Y. Nedellec, A. Kreisz, N. Donzel, J. Rozière, D. J. Jones, F. Millo, A. Piu
- **Analysis of a HT-PEMFC range extender for a Light Duty Full Electric Vehicle (LD-FEV)**, Federico Millo, Sabino Caputo, Alessandro Piu
- **Catalyst distribution effects on performance and durability of HT PEMFC**, Uktam R. Salomov, Eliodoro Chiavazzo and Pietro Asinari
- **MEA development, optimisation and testing, stack assembly and testing**, A. Morin, C. Nayoze, N. Donzel, D. Jones, J. Rozière, Y. Nedellec, W. Kerkhof, M. Smit

PARTICIPATION IN INTERNATIONAL CONFERENCES

The partners have attended prominent international conferences, workshops and symposia and have presented the following contributions to oral and poster programmes:

1. **European Hydrogen Energy Conference,** Seville, Spain, March 2014.
   Novel materials for a HT-PEMFC stack for operation as automotive range extender, Y. Nedellec[1], A. Kreisz[1], N. Donzel[1], J. Rozière[1], D. J. Jones[1], A. Morin[2], C. Nayoze[2], F. Alcaide Monterrubio[3], M. Smit[4], P. Asinari[5], U. Salomov[5], A. Piu[6], [1]CNRS - Institut Charles Gerhardt, Agrégats Interfaces et Matériaux pour l'Energie - UMR 5253 CNRS – France, [2]CEA - Commissariat à l'Energie Atomique et aux Energies Alternatives - LITEN – France, [3]CIDETEC – Spain, [4]Nedstack – Netherlands, [5]Politecnico Di Torino – Italy, [6]CRF – Centro Ricerche FIAT – Italy.
2. **European Hydrogen Energy Conference,** Seville, Spain, March 2014.
   Multi-scale modelling to boost fuel cell performance: From pore-scale simulations to better efficiency and durability, U. Salomov, P. Asinari, Politecnico di Torino, Italy
3. **International Society of Electrochemistry,** Lausanne, Switzerland, August-September 2014.
   Performance of HT-PEMFC on operation as automotive range extender, Y. Nedellec, A. Kreisz, N. Donzel, D. Jones, J. Rozière, CNRS Montpellier
4. **8th International Conference on Sustainable Energy & Environmental Protection (SEEP 2015),** Paisley, Scotland, 2015.
   *Analysis of a HT-PEMFC range extender for a Light Duty Full Electric Vehicle (LD-FEV).*F. Millo, S. Caputo, A. Piu, Politecnico di Torino, Centro Ricerca FIAT
5. **Electrolysis and Fuel Cell Discussions Challenges Towards Zero Platinum for Oxygen Reduction (EFCD2015),** 13-16 September 2015, Palais des Congrès, La Grande Motte, France
   Catalyst distribution effect on performance and durability of HTPEMFC, Uktam R. Salomov, P. Asinari, Politecnico di Torino
6. **Electrolysis and Fuel Cell Discussions Challenges Towards Zero Platinum for Oxygen Reduction (EFCD2015),** 13-16 September 2015, La Grande Motte, France
   New low Pt Loading Fuel Cell Cathode Architecture made of Self-standing Arrays of Electrocatalyst Nanotubes, A. Morin, N. Pau, S. Galbiati, O. Marconnot, and D. Buttard, CEA LITEN
7. **Electrolysis and Fuel Cell Discussions Challenges Towards Zero Platinum for Oxygen Reduction (EFCD2015),** 13-16 September 2015, La Grande Motte, France

Cross-Linked Phosphoric Acid Doped PBI Membranes and Their Performance on Operation As Automotive Range Extender, N. Donzel, A. Kreisz, Y. Nedellec, D. J. Jones, J. Rozière, CNRS Montpellier

8. **International Society of Electrochemistry Annual Meeting**, September 2014, Lausanne.
   Performance of HT-PEMFC on operation as automotive range extender, Y. Nedellec, A. Kreisz, N. Donzel, D. J. Jones, J. Rozière, CNRS Montpellier
9. **Conference "Matériaux 2014",** November 2014, Montpellier
   Nouvelles membranes polybenzimidazole pour pile à combustible à haute température, A. Kreisz, N. Donzel, Y. Nedellec, D. J. Jones, J. Rozière, CNRS Montpellier
10. **ECS Meeting on Electrochemical Energy Conversion and Storage**, 27 July 2015, Glasgow
   Cross-linked phosphoric acid PBI membranes and their performance on operation as automotive range extender, N. Donzel, A. Kreisz, Y. Nedellec, D. J. Jones, J. Rozière, CNRS Montpellier

PRESENTATIONS AT FCH-JU REVIEW DAYS

1. **FCH-JU programme review days, Brussels, 11-12 November 2013**
   - Poster presentation of the ARTEMIS project, Y. Nedellec, CNRS Montpellier, France
2. **FCH-JU programme review days, Brussels, 10-11 November 2014**
   - Oral presentation of the ARTEMIS project, Y. Nedellec, CNRS Montpellier, France
3. **FCH-JU programme review days, Brussels, 17-18 November 2015**
   - Poster presentation of the ARTEMIS project, D. Jones, CNRS Montpellier, France

JOURNAL PUBLICATIONS

The Consortium has published the following publications in international scientific journals:

1. **Pore-scale modeling of fluid flow through gas diffusion and catalyst layers for high temperature proton exchange membrane (HT-PEM) fuel cells**, U. R. Salomov, E. Chaivazzo, P. Asinari, Comp. Math. Appl., (2013), http://doi.org/10.1016/j.camwa.2013.08.006
2. **Gas-dynamic and electro-chemical optimization of catalyst layers in high temperature polymeric electrolyte membrane fuel cells**, Uktam R. Salomov, Eliodoro Chiavazzo, Pietro Asinari, International Journal of Hydrogen Energy, 40, 5425, (2015) http://dx.doi.org/10.1016/j.ijhydene.2015.01.059

DISSEMINATION SUPPORT MATERIALS

Various supports were developed to ensure a visual identity for the ARTEMIS project. These included a project logo, and a powerpoint presentation template, used for project presentations at conferences, workshops, progress meetings etc.

4.2 FUTURE DISSEMINATION PLANS

The consortium will be engaged in conducting further activities dedicated at disseminating the project results, in particular through journal publications. The following measures are planned to follow up the project:

ARTEMIS WEBSITE

The ARTEMIS website will kept as an information source of the activities performed in the project. The website will also continue to receive and upload the abstracts of papers from the project.

JOURNAL PUBLICATIONS

Future academic articles and reports are an important component in the continuation of communicating the results from the research undertaken. Partners will target specifically relevant well-recognized academic journals for the future publications. The following manuscripts have bee submitted or are in preparation:

1. Analysis of a HT-PEMFC range extender for a Light Duty Full Electric Vehicle (LD-FEV). F. Millo, S. Caputo, A. Piu, International Journal of Hydrogen Energy (submitted).
2. Pore- and Macro-scale Simulations of High Temperature Proton Exchange Fuel Cells - HTPEMFC - and Possible Strategies for Enhancing Durability. Uktam R. Salomov, Eliodoro Chiavazzo, Pietro Asinari, International Journal of Hydrogen Energy (submitted).
3. Modeling and experimental studies to improve high temperature PEM fuel cell durability for automotive range extender application. Uktam R. Salomov, Eliodoro Chiavazzo, Pietro Asinari, Arnaud Morin, Cristine Nayoze, Federico Millo, Sabino Caputo (in preparation).
4. Effect of catalyst distribution on transport properties of HT PEMFC. Uktam R. Salomov, Eliodoro Chiavazzo, Pietro Asinari (in preparation).
5. Cross-linked and reinforced polybenzimidazole with high level of acid doping for high temperature PEMFC, Aurelien Kreisz, Nicolas Donzel, Yannig Nedellec, Deborah Jones, Jacques Rozière (in preparation)

PATENTS

A patent is in preparation at CEA on the fabrication of high temperature MEAs.

PHD THESIS

PhD thesis of Aurélien Kreisz, Université de Montpellier.

FUTURE CONFERENCE PRESENTATIONS

ARTEMIS partners will continue to disseminate the project results in relevant conferences, including:

1. **Fuel Cells 2016 Science & Technology conference, A Grove Fuel Cell Event, Glasgow, 13-14 April 2016**
   - *Novel polymer nanofibre reinforcements for robust and durable fuel cell membranes*. D. Jones, J. Rozière, N. Donzel, A. Kreisz.
2. **STEPI 10 – Polyimides and high performance polymers, Montpellier, 6-8 June 2016**
   - *Novel polymer nanofibre reinforcements for robust and durable fuel cell membranes*. D. Jones, J. Rozière, N. Donzel, A. Kreisz.

4.3 POTENTIAL IMPACT AND EXPLOITATION OF FOREGROUND

POTENTIAL IMPACT

CNRS has developed a proton conducting membrane suitable for high temperature PEMFC. It differs from currently available HT PEMFC membranes in that although it is highly acid doped due to the sol-gel type preparation process, it is mechanically strong, since the polymer is partially cross-linked, and the membrane is reinforced using an electrospun nanofibre mat of cross-linked polymer. The process is scalable, since in ARTEMIS CNRS has scaled up the preparation of the reinforcement materials and the membranes and produced ca. 40 membranes of area 325 cm^2. The membranes have been validated in large size MEAs and a short stack, with operation on a variable load range extender protocol (1000 hours to end of test with single cells). The foreground might be exploited by an industry producing membranes by a casting process. Further research is required to transfer the current batch process to a continuous processing line. The targeted application area is in MEAs for high temperature PEMFC, although application in MEAs for electrochemical H_2 compression and in H_2 purification/pumping is also envisaged. Application areas of HT PEMFC include APUs, range extenders and micro-CHP. During the course of ARTEMIS, CNRS has extended its knowledge and expertise on the fabrication of membranes for high temperature PEMFC. This improved expertise will be used for CNRS' own development and research activities and in future national and European projects. The membranes which have been developed exceed the best commercial state of the art and have enabled, along with the advances at CEA in electrode development, to surpass the commercial state of the art for HT PEMFC performance.

CIDETEC has developed an anode catalyst suitable for high temperature PEMFC. It differs from currently available HT-PEMFC catalysts in the support of metal nanoparticles. The catalyst is currently manufactured by a batch process, and it is scalable, since in ARTEMIS CIDETEC has carried out the scale-up of the manufacturing process from 0.4 g to tenfold the initial batch. The catalyst has been tested in 25 cm^2 high temperature single cell fed with H_2 and carbon monoxide (0.5 – 5 %). The foreground might be exploited by transferring the technology to an industrial catalyst manufacturer. The targeted application area is in high temperature PEMFC (APUs, range extenders and micro-CHP), although application in MEAs for low temperature PEMFC is also envisaged. In the framework of ARTEMIS, CIDETEC has gained knowledge and expertise on catalyst manufacturing and characterization using alternative supports intended for high temperature PEMFC. The project allows CIDETEC to increase its expertise and offer in the field of PEMFC fuel cells R&D, which will be used for internal development, and research activities in future national and European projects. The catalyst developed shows increased tolerance to high concentrations of CO compared to state of the art Pt/C catalyst, and work is in progress to transfer the know-how to an industrial catalyst manufacturer, which produces catalysts for the targeted applications.

CEA has developed gas diffusion electrodes for high temperature PEMFC which are made by coating. The process is scalable, since in ARTEMIS CEA has manufactured several tens of electrodes of around 600 cm^2. The electrodes have been validated in 25 cm^2 single cell fixture with operation on a variable load range extender protocol (2000 hours to end of test) but also in large size MEAs and in a short stack. The foreground might be exploited by transferring the technology to an industrial. The targeted application area is in MEAs for high temperature PEMFC, although application in MEAs for electrochemical H_2 compression or H_2 purification is also envisaged. During the course of ARTEMIS CEA has acquired knowledge and expertise on the fabrication and characterisation of electrodes and Membrane Electrode Assemblies for high temperature PEMFC. The project allows CEA to increase its expertise and offer in the field of PEM fuel cells research and development. This new expertise will be used for CEA's own development and research activities and in future national and European projects. The MEA which has been developed exceed the best commercial state of the art and work is in progress in order to transfer the technology to an industrial partner.

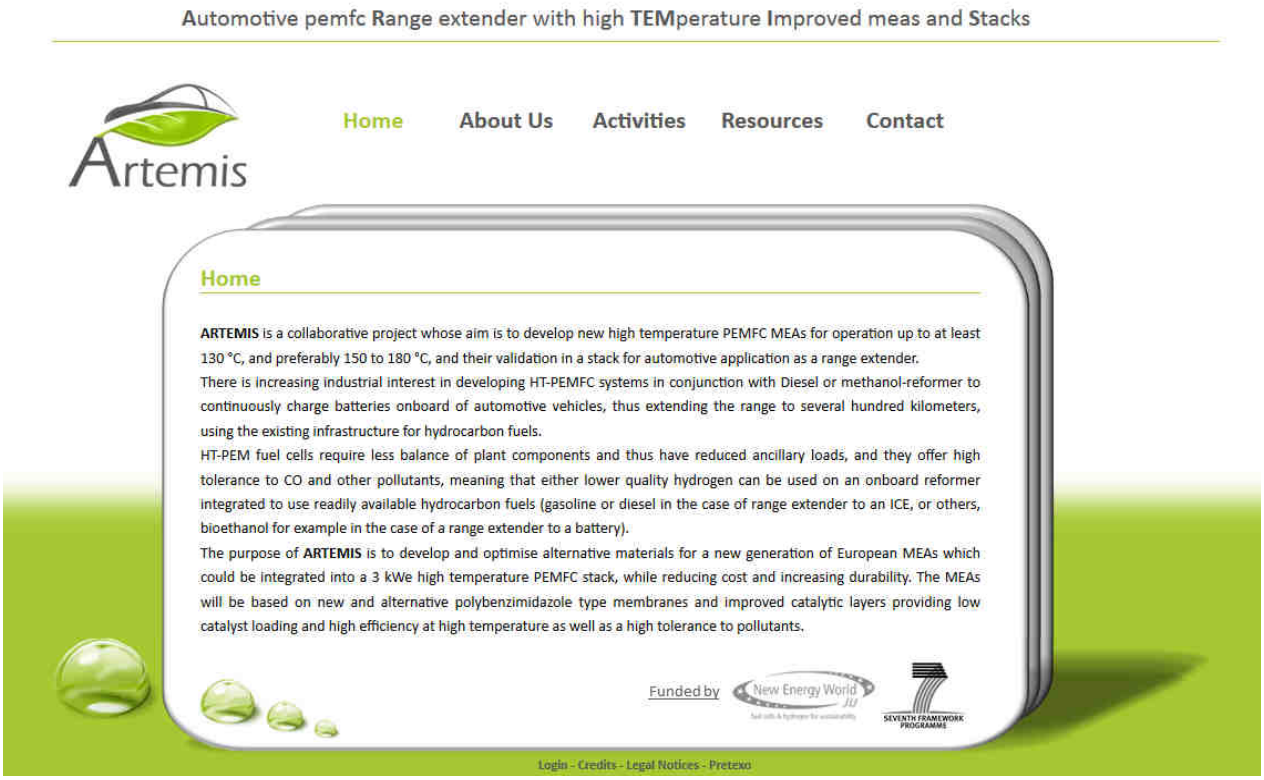
Nedstack as an independent fuel cell stack manufacturer is always looking toward stack improvements, innovation and new technology to maintain and strengthen its market position. The HT-PEMFC technology developed within the ARTEMIS project is of great commercial interest, since it eliminates the need for purification of CO from less pure hydrogen sources. From Nedstack's point of view, this technology may be implemented not only in range extenders for automotive applications, but also in other applications, such as telecom. Since Nedstack's business model is based on selling stacks to system integrators, close collaboration needs to be established with system integrators in order to design, develop and commercialise the appropriate balance-of-plant, for each specific application.

In the course of the ARTEMIS project, significant progress has been made towards increased durability and performance of HT-PEMFC components and stacks, together with reduced production costs for MEAs and cell plates. Nedstack will now continue to develop the business case for HT-PEMFC as part of their products. The results of the evaluation of financial aspects together with the current knowledge of component cost and lifetime, will allow assessment of whether commercialisation is currently viable or whether the HT-PEMFC stack still need further development or improvement. In case the initial rough business case indicates a good market position, a more detailed viability study will be started and the route towards commercialisation will be determined. As part of the viability study, the market for this technology will be re-evaluated. The in-house production processes for the stack components will be screened, fine-tuned and modified where needed. Quality criteria will be established for stack components as well as the final product. Negotiations with suppliers will be started. Finally a range of stack sizes, based on power output, will be determined, based on the expected technology application. In case further development of components is needed, Nedstack may continue the close collaboration with the ARTEMIS project partners, and thus take advantage of the knowledge and experience gained in this project. Specific agreements may be set up between partners to work on specific components and safeguard proprietary information. Once the final HT-PEMFC product has been fully completed, configured and determined, Nedstack will start a publicity campaign of its HT-PEMFC technology and together with the system integrator will work towards a sales strategy to final customers.

EXPLOITATION OF FOREGROUND: ARTEMIS has led to five exploitable foregrounds on: high temperature membrane, anode catalyst, gas diffusion electrode, high temperature cell plates and high temperature stack, HT PEMFC MEA configuration.

EXPLOITATION OF FOREGROUND IN FUTURE COLLABORATION BETWEEN PARTNERS: Discussions have been initiated on how to use best the results of ARTEMIS in a next stage of development dedicated principally to pilot level scale-up/manufacture, materials and processing optimisation, cell and stack development, for the automotive sector. A future opportunity for funding is sought under FCH 2 JU.

5. ADDRESS OF THE PROJECT PUBLIC WEBSITE



Automotive pemfc **R**ange extender with high **TEM**perature **I**mproved meas and **S**tacks

Home

ARTEMIS is a collaborative project whose aim is to develop new high temperature PEMFC MEAs for operation up to at least 130 °C, and preferably 150 to 180 °C, and their validation in a stack for automotive application as a range extender.

There is increasing industrial interest in developing HT-PEMFC systems in conjunction with Diesel or methanol-reformer to continuously charge batteries onboard of automotive vehicles, thus extending the range to several hundred kilometers, using the existing infrastructure for hydrocarbon fuels.

HT-PEM fuel cells require less balance of plant components and thus have reduced ancillary loads, and they offer high tolerance to CO and other pollutants, meaning that either lower quality hydrogen can be used on an onboard reformer integrated to use readily available hydrocarbon fuels (gasoline or diesel in the case of range extender to an ICE, or others, bioethanol for example in the case of a range extender to a battery).

The purpose of **ARTEMIS** is to develop and optimise alternative materials for a new generation of European MEAs which could be integrated into a 3 kWe high temperature PEMFC stack, while reducing cost and increasing durability. The MEAs will be based on new and alternative polybenzimidazole type membranes and improved catalytic layers providing low catalyst loading and high efficiency at high temperature as well as a high tolerance to pollutants.

http://www.artemis-htpem.eu

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