PROJECT FINAL REPORT

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List of Acronyms

AFM	atomic force microscopy
AUTOSTACK	acronym of EU Project: Automotive High Temperature Fuel Cell Membranes
BOT	begin of test
CFD	computational fluid dynamic
CRT	Cell reversal tolerance
CV	cyclic voltammetry
DM	Diffusion medium
EDX	energy dispersive X-ray spectroscopy
EW	Equivalent weight
fcc	face-centered cubic (crystal structure)
FC-DLC	Fuel cell dynamic load cycle
FIB	Focussed ion beam
GDL	gas diffusion layer
IRfree	voltage without voltage loss of ohmic resistance
KB	Ketjenblack (carbon black from Akzonobel)
MC	Monte Carlo simulation
MEA	membrane electrode assembly
MPL	micro porous layer
OCV	open-circuit voltage
OEM	original equipment manufacturer
ORR	Oxygen reduction reaction
PE(M)FC	polymer electrolyte membrane fuel cells
PEM	polymer electrolyte membrane fuel cells
PNM	pore network model
PTFE	polytetrafluoroethylene
PFSA	perfluorosulfonic acid
Pt	platinum
RH	relative humidity
RHE	Reversible hydrogen electrode
P1	1st reporting period
RT	room temperature (23°C)
SEM	scanning electron microscopy
SGL	SGL carbon group
TEM	Transmission electron microscopy
UV	ultra-violet
XPS	X-ray photoelectron spectroscopy
XRD	X-ray diffraction
XRF	X-ray fluorescence

4.1   Final publishable summary report

Executive Summary

The main objectives of IMPACT were:

- Improvement of lifetime of low loaded MEAs (goal **5,000 hours dynamic operation** in stack and degradation rates below **10 µV/h**, with a Pt loading of **0.2 mg/cm²**)
- Development of durable low loaded MEAs for automotive applications with single cell performances achieving **1 W/cm²** at 670 mV cell voltage

In order to achieve these goals substantial effort was made in order to increase the chemical and electrochemical stability of the **catalysts**, in a wide range of operating conditions. To increase cell performance thin **10 µm thick AQUIVION® PFSA membranes** have been developed. Moreover, the new membranes have been chemically stabilized to assure high resistance against corrosion and reduced hydrogen cross-over. Further effort has been dedicated to the **development of ionomers** for the catalyst layers. In the context of catalyst layer development the introduction of stabilized, i.e. highly graphitized carbon supports lead to a significant improvement of CCM durability in in accelerated stress tests.

The basis for the research was extensive single cell and stack testing at the industrial and research partner`s facilities and detailed analysis and evaluation of degradation. The project approach was iterative material development and testing (7 generations). Thanks to detailed ex-situ and in-situ analysis of the developed materials a better understanding of degradation mechanism could be obtained.

In the course of the project the Pt loading was reduced from 0.6 mg cm^{-2} to 0.2 mg cm^{-2}. The achieved durability still needed to be improved. While in 500 – 1000 h single cell tests promising irreversible degradation rates of 10 - 60 µV/h were measured, the final durability stack test with the same optimised MEAs had to be terminated after only 1700 h due to some unexpected and unexplained issues with the MEAs. A power density up of **1.0 W cm^{-2}** at 1.8 A cm^{-2} could be reached at **0.2 mg cm^{-2}** total loading (0.15 mg cm^{-2} cathode loading) corresponding to the state-of-the-art performance at such low loading. According to our study on the influence of Pt loading on performance and degradation the threshold Pt loading below which a significant drop of performance is expected corresponds to 0.2 mg cm^{-2}.

A dedicated cost model and a life cycle analysis have been developed to assess the sensitivity of stack cost on performance, Pt loading and degradation.

Moreover, an innovative model of a PEMFC cathode has been proposed, coupling a pore network model (PNM) for describing the two-phase transfers in the diffusion medium (DM) of a GDL to a continuum approach in the MPL, catalyst layer and membrane. The advantage of the PNM used here is to allow the modelling of the liquid water formation by condensation in the DM using a state-of–the art condensation algorithm.

In the course of the project two international workshops and a summer school have been organized.

Project Context and Objectives

Passenger vehicles powered by fuel cells offer significant advantages over today's internal combustion engines (ICEs) and offer a driving range similar to ICEs. Fuel cell stacks generate electricity without combustion, producing zero-emissions and little, if any, noise or vibration. Fuel cells have few efficiency limitations imposed on them in comparison to traditional combustion engines, and consequently have very high efficiencies in converting chemical energy to electrical energy. Therefore, many large automotive companies have committed themselves to a long term development for automotive fuel cell power trains and the necessary parallel implementation of a hydrogen fuelling infrastructure. Prominent European examples for the government support of this strategy are the German National Innovation Program and the Initiative H_2-Mobility to implement hydrogen filling stations.

This project contributes to these goals by addressing the important topic of cost reduction of polymer electrolyte fuel cells (PEFC) at high performance and with sufficient durability. The automotive fuel cell application demands the most stringent requirements in terms of performance, cost-effectiveness and permitted mass/volume of stacks and systems. The following requirements have been summarized numerously by the OEMs: The stack durability (lifetime) of a passenger car should be about **5,000 – 5,500 h** (20,000 h for buses) **of operation** with less than **50** to **75 mV** degradation at a given current density. In addition, **300,000 large load cycles** (possibly as low as 30,000 with hybridization with battery) are encountered which lead to formation and reduction of Pt surface oxides at the cathode, possibly Pt dissolution and re-deposition. Furthermore, **30,000 start-up/shutdown** cycles are typically required in a normal car operation leading to local potentials of 1.5 V and above at the cathode electrode, which can lead to rapid corrosion of carbon support. Around **2,500 subfreezing starts** with transitions through frozen and liquid water products are required in the colder climates[2]. An aspect of paramount importance is the **cost reduction requirement** in which the MEA dominates and is associated with the precious metal content (primarily platinum), membrane and other components. Moreover, in the scenario of mass volume production of about 500,000 stacks production per year for automotive applications, the precious metal content of the electrodes and MEAs will dominate stack (54%) and system costs (27%). Therefore, it is imperative to reduce the precious metal content in the MEAs to achieve the goals of commercial market introduction in car series production.

A target **Pt loading of <0.2 $g_{Pt}kW^{-1}$** and a power density > 1 **Wcm^{-2}** at e.g. cell voltages of **0.67 V** (to allow heat rejection) have been developed and widely accepted as the required goals for the automotive application. These targets correlate to a cathode kinetic mass activity of > **0.44 Amg^{-1}_{Pt} @ 900 mV** (Pt cost), which in general represents a 3-4 fold increase in activity over a baseline Pt/C catalyst.

Nevertheless, a **trilemma** is present in the state-of-art PEFCs (Figure 1). The reduction of precious group metal content in the cells exacerbates the concomitant requirement to maintain the performance level of the cell and also of the durability (lifetime) of at least 5,000 h. Therefore development of new materials and improved membrane electrode assemblies is still required to enable cost, performance and lifetime targets to be attained.

2 F. Wagner, H. Gasteiger, R. Makharia, S. G. Yan "Catalyst Development Needs and Pathways for Automotive PEM Fuel Cells ", ECS 210th Meeting, Abstract 0433.pdf
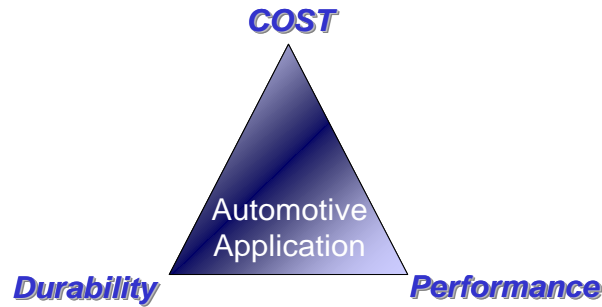
Figure 1: Trilemma of PEFC automotive application: The concurrent decrease of noble metal loading to <0.2gkW⁻¹Pt and the maintenance of performance levels and durability.

Accordingly, the main objective of IMPACT was to increase the life-time of low Pt loaded (0.2 mg cm^{-2}) membrane –electrode-assemblies for automotive applications without compromising the cell performance. The economic requirements to reduce Pt loading lead to the challenge to maintaining durability and performance, an aspect which has not been addressed sufficiently in public projects and studies so far. In line with these targets the main project goals can be summarized as:

- Improvement of lifetime of ultra-low loaded MEAs (goal 5,000 hours dynamic operation in stack and degradation rates below **10 µV/h**, with a Pt loading of **200 µg/cm²**)
- Development of durable low loaded MEAs for automotive applications with single cell performances achieving **1 W/cm^2** at 670 mV cell voltage

In order to achieve these two key goals particular effort was devoted to understand the fundamental degradation processes of at low loadings and the relation between degradation, life time and Pt-loading. Furthermore, improvement of components under different operating conditions and demonstration of improved lifetime have been undertaken. The basis for the research was extensive iterative material development and testing (Figure 2) at the industrial and research partner`s facilities and detailed analysis and evaluation of degradation processes and their importance for fuel cell performance loss.
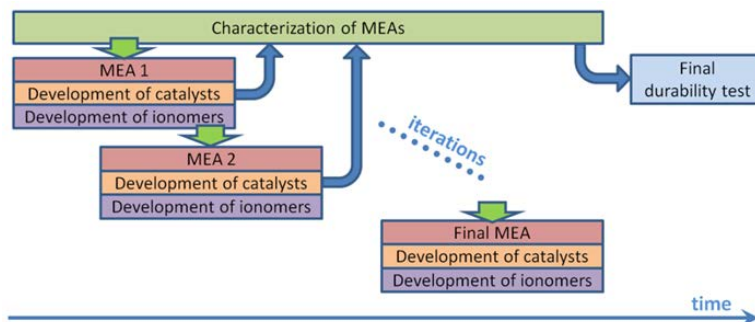

Figure 2: Simplified Gantt-Chart of IMPACT demonstrating the iterative approach of MEA development

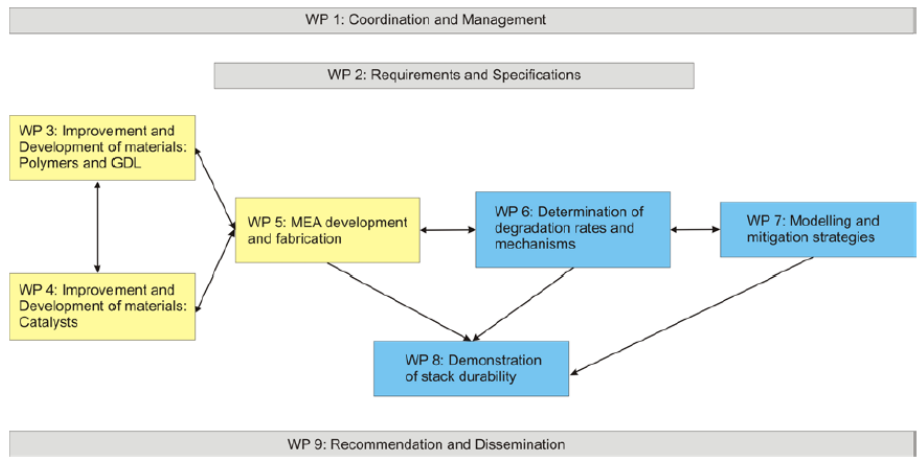The project was composed of the work packages structure summarized in Figure 3.

Figure 3: Schematic representation of the structure of IMPACT illustrating the work activities and the interaction of the work packages.

Description of the main S&T Results

In the following the achievements of the technical work packages WP2 – WP8 are outlined.

WP2: Requirements and Specifications

Within WP2 MEA (CCM and GDL), cell and stack hardware as well as test procedures used for performance and durability testing have been defined. Furthermore, a dedicated cost analysis for low loaded MEAs was developed taking into account the requirements of the automotive application the IMPACT was aiming at. Thus, this WP was closely related to the WPs 3 to 5 dedicated to material development (membrane and GDL, catalyst, MEA), furthermore to the WPs 6 and 8, where fuel cell and stack tests have been performed. It also supported WP7 activities, as the modelling activities there require basic conditions and material properties as input.

Within to course of the project the set of operating conditions and test procedures was continuously extended and updated to reflect progress in relevant project-external activities. Furthermore, a sensitivity analysis of electrode parameters was performed in order to investigate the influence of selected electrode parameters on the cell performance. The objective of this sensitivity analysis was to support the MEA development and fabrication (WP5) by identifying optimization potentials for this purpose.

Computational fluid dynamics (CFD) models were used to perform the sensitivity analysis. In total, eight electrode parameters were varied. The impact of the electrode parameter variation on the cell performance was assessed by comparing the cell voltage against reference cases. The knowledge gained can be used to specify MEA parameters needed for achieving the automotive target values. Thus, the simulation activities carried out can contribute and support the development of durable ultra-low loaded MEAs for automotive applications also beyond IMPACT.

Three relevant cost models were developed within the cost assessment effort. Out of these, both the detailed production cost model and the lifecycle cost model provide valuable economic information which can be used to guide further development. Notably, the lifecycle cost model employed here allows insight on new aspects which were not dealt with in literature so far. The simplified cost model provides a convenient tool for fast assessment of the impact of possible platinum price spikes. Congruently, the economic models predict a beneficial effect for the introduction of the MEAs resulting from the project into the market, even though the original technical performance and degradation targets could not be fully met.

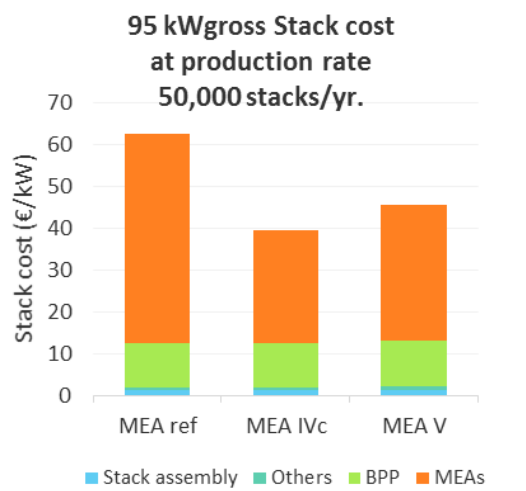**Figure 4: Stack cost for different MEAs according to the developed cost model.**

In total, for WP2 both the original targets as specified in the DoW were met and the additional modelling efforts agreed during midterm reviewing were successfully conducted. Interim delays occurring with respect to the delivery of results had no significant influence on the final success of the work performed.

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WP3: Improvement and Development of Materials: Polymers and GDL

Work Package 3 is focused on the development and improvement of materials, in particular membranes, ionomer dispersions for electrode and GDLs. The development activity of membranes and ionomer dispersions was led by SLX whereas GDL improvement was managed by CEA. In the frame of this work package and in view of the better understanding of the structure-performance relationship of developed materials, a relevant role was also played by the advanced characterization carried out by UAES.

Material Development

With the aim to achieve the best utilization of Pt catalyst and thus reducing its loading while maintaining performance and durability high enough to meet the requirements of automotive industry, SLX, starting from the reference materials developed and made available to the partners two generations of membranes and ionomer dispersions (Figure 5).
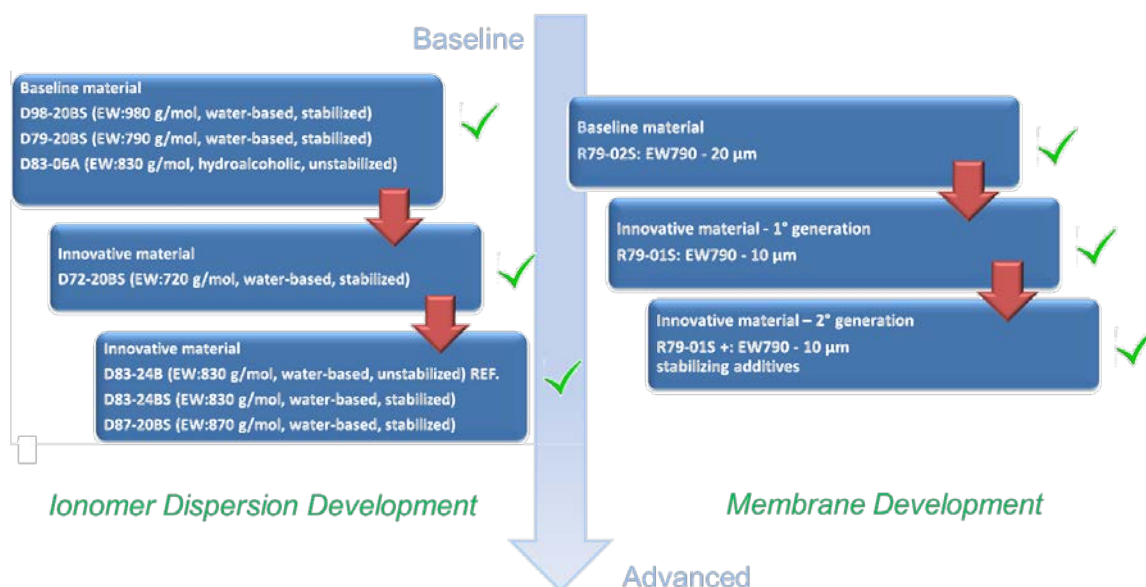

Figure 5: Pathway of ionomer dispersions (left) and membrane (right) development at SLX

Membranes developed are based on Aquivion® PFSA ionomer and are reinforced with an internal support constituted by expanded PTFE in order to extend lifetime by increasing their resistance towards mechanical stress induced by dry-wet cycles during operations. With the aim to get the electrochemical performance high enough also at low Pt-loading, during the project, membrane thickness was reduced from 20 to 10 μm maintaining the ionomer equivalent weight (EW) of 790 g/mol corresponding to an ion-exchange capacity of 1.26 mmol/g.

In the second generation of improved membrane, innovative radical scavengers able to reducing polymer chemical degradation triggered by radicals produced within the fuel cell hence increasing membrane durability without affecting electrochemical performance have been introduced.

Dispersions have been also iteratively improved by changing the polymer EW and related properties (crystallinity, ion-exchange capacity, hydrophilic/hydrophobic balance, water uptake and gas permeability) starting from commercially available Aquivion® PFSA dispersions.

Testing carried out by partners in the frame of Impact pointed out that ionomer in electrode suffers of instability, leading, especially during dry-wet cycles, to a sort of "restructuration" within the catalytic layer when the EW is below the threshold of 800 g/mol and thus for the final iteration water-based Aquivion® PFSA dispersion having physical properties and polymer composition finely tuned to achieve good stability and good electrochemical performance have been developed.

Aquivion® PFSA dispersion D83-24BS and R79-01SX+ membrane developed and improved along the project has been selected to be used to prepare MEAs for the final stack.

Material Characterization

The GDL/MPL, the membrane and the electrodes have been analysed by material-sensitive atomic force microscopy (AFM) as well as optical spectroscopic tools, mainly on freshly microtome cut cross-sections of MEAs. AFM is especially suited for analysis of the ionomer because investigation can be performed at controlled high humidity and at elevated temperatures, close to operational conditions.

Membranes

For the analysis of the hydrophilic/hydrophobic phase structure in the ionomer, two methods were used, the adhesion force (pull-off force of tip from surface) mapping and the contact current measurement. In the latter, the current measured in tapping mode was averaged by lock-in amplifier during contact of tip and sample. Using contact current mapping the differences in size and geometry of the water-rich, conductive areas in Nafion with long side chains in the molecule and Aquivion PFSA with short side chains in the molecule were found. In adhesion mappings on ionomer, the ionomer phase structure with low-adhesive, water-rich hydrophilic, ionically conductive areas and high-adhesive dry hydrophobic backbone-rich areas could be discerned.

Electrodes

In adhesion force mappings on electrodes, the ionomer can be discerned by its high adhesion to the AFM tip from carbon and platinum. Due to their very low adhesion they appear black in the measurements. In high-resolution measurements with a pixel size < 1 nm and a nominal tip radius below 1 nm, the ionomer coverage around Pt/C agglomerates was studied. At cross-sections of 1 μm^2 of the anode and cathodes the thicknesses of all layers were determined.

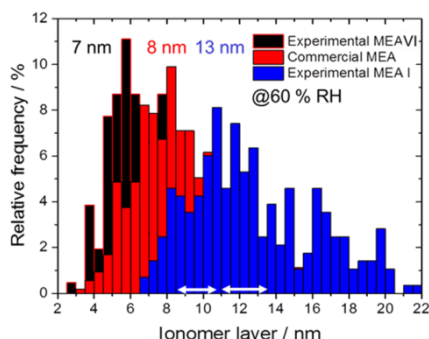
Figure 6: Distribution of ionomer layer thicknesses in Nafion based non-automotive commercial reference MEA(Alfa-Aesar), project reference MEA 1 and advanced IMPACT MEA 4a, measured at 60 % RH.

As layer thickness, the peak position of log-normal fits to the resulting histograms was taken. The distribution ranged from minimal 3.5 in case of Aquivion PFSA and 4.5 in case of the Nafion-based

MEA to a maximal layer thickness around 20 nm. As visible in Figure 6, the distribution and the peak positon depends on preparation, ranging from 7-13 nm in advanced IMPACT MEA 4a, Nafion-based commercial reference MEA (a non-automotive MEA sourced commercially from Alfa Aesar), and project reference MEA 1, respectively. In Figure 7, the ionomer layer thickness for the IMPCT MEA generations is given. The layer thickness decreased with proceeding generations with the exception of the final MEA 5.
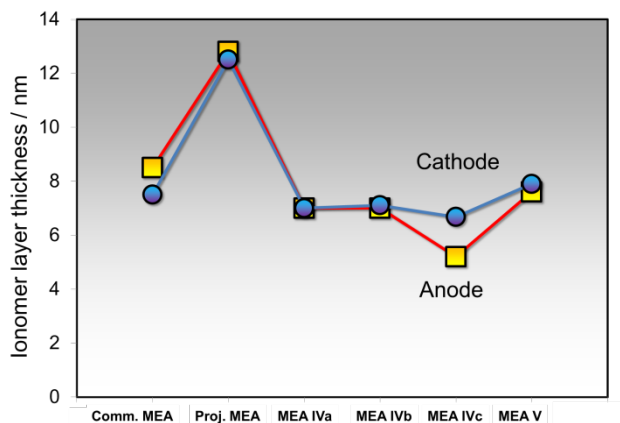

Figure 7: Ionomer layer thickness of MEA generations at anode and cathode, normalized to project reference MEA, measured at 60 % RH.

For investigation of the conductivity of such ultra-thin layers present in the electrode, self-assembled films were prepared. Through-layer steady-state conductivity of the resulting lamellar films started above a film thickness of more than 10 nm. Below this thickness, the ionic conductivity was present in a water layer below the ionomer deposited on a hydrophilic surface as Pt catalyst on carbon. Since a significant fraction of the ionomer layers in the electrodes has a thickness below 10 nm, at least a part of them with lamellar might have not through-layer conductivity.

GDL/MPL
The surface of improved IMPACT SGL BC25 GDL was investigated after artificial aging with H_2O_2 and improved durability concerning loss of PTFE/increase of conductive area was measured over standard material.

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WP4: Improvement and Development of Materials: Catalysts

The general objectives of this work package was the enhancement of the PEM fuel cell reaction rates (anodic and cathodic) with respect to the state of art electro-catalysts at conventional (80°C) and high temperatures (110°C) at a low relative humidity (33% RH). The aim was to increase the catalyst chemical and electrochemical stability in a wide range of operating conditions.

To achieve such objectives, research activities dealing with screening procedures and characterization of catalysts by ex-situ (XRD, TEM, EDX, XRF) and in situ (CV, polarisation, AC-impedance) techniques were undertaken.

The potentialities of new developed materials were assessed through a screening via half-cell and single cell experiments. The half-cell experiments were carried out in the low temperature range in the presence of liquid electrolytes (< 80°C) to determine electro-kinetic parameters and by using proper catalyst/ionomer assemblies in order to investigate the electrochemical active area, corrosion/degradation resistance. Single cell testing was conducted at different temperatures, pressure and humidification operating conditions appropriate for automotive applications as defined in WP2.

Anode catalyst development was a relevant aspect of the project since lowering the MEA catalyst loading led to an increase of sensitivity to contaminations and to surface area loss.

The anode catalyst was designed to provide increased stability and higher tolerance to low levels of impurities and to allow for an effective decrease of the precious noble metal loading. The activity carried out in the first period of WP4 was dealing with a comparison of Pt/C catalysts prepared using different supports and their comparison with benchmark commercial catalysts.

The poisoning of the Pt/C anode electrocatalyst by trace levels of CO is a known problem, the resulting limitation of the hydrogen oxidation reaction (HOR) functionality ultimately reducing the performance of the overall membrane electrode assembly (MEA). Thus, the consortium has been actively looking at a range of catalyst which are tolerant to low levels of CO and particularly, catalysts which are free from Ru given the known problems associated with Ru dissolution and its subsequent ability to poison the fuel cell cathode.

The general objectives for the anode development in this work package were achieved. These regarded an enhancement of the anodic reaction rate and stability for the Pt/KB catalyst obtained by a carbothermal procedure versus the conventional Pt/Vulcan reduced in a hydrogen stream. The comparative data recorded in a PEM single cell at conventional (80°C) and high temperatures (110°C) at low relative humidity (33% RH) indicated similar performance for the two catalysts at conventional conditions in the range of technical interest, but a better performance for the Pt/KB at high temperatures and low RH. The latter conditions were particularly useful for automotive application. Notably, the Pt/KB catalyst showed a better electrochemical stability as assessed by both in-situ and ex-situ investigations. The new developed catalyst was assessed in the presence of the Aquivion membrane and ionomer developed in WP3 with promising results.

The oxygen reduction reaction (ORR) is the rate-determining step in polymer electrolyte membrane fuel cells; moreover since the oxygen reduction catalyst operates at high potentials, cathode corrosion represents a significant issue. The cathode catalysts thus deserve special attention both with regard to the electrocatalytic activity and resistance to degradation. The activity dealing with the catalyst development for the cathode regarded the implementation of alloying and crystallographic structure characteristics as well as the use of alternative supports to achieve superior electrocatalytic activity and stability.

In this WP, most of the activity was addressed to alloying Pt with transition metals and tailoring the particle size. Practical procedures, such as pre-leaching of the alloy in an appropriate acid, to increase the Pt in the external layers of supported nanoparticles were developed. Mass activity values were determined in situ using practical catalysts loadings to get insights into the possible achievable MEA performance.

JMFC also had a broad range of catalyst development activities which were already in progress outside of the IMPACT project. Down-selected catalyst candidates which showed significant promise were brought into the project for further development and for integration into novel MEA designs being developed in IMPACT. A Pt/C electrocatalyst with enhanced stability was combined with Solvay's 790EW ionomer and the new, 10 micron PFSA membrane to provide a next-generation MEA for the project.

Also a non-Co binary alloy layer was developed such that the required reduction in cathode loading was achieved without compromising the overall activity of the PEMFC MEA. Optimisation work on the deposition chemistry, and translation of the catalyst to the more stable carbon of the Pt/C work was actively carried out within this programme.

Carbon-supported PtNi and PtCo cathode electro-catalysts were developed at CNR-ITAE to evaluate their performance and stability as cathodes in a polymer electrolyte membrane fuel cell (PEMFC) under various conditions of operating temperature and pressure. Sulphite complex and acid formic-based preparation routes were used. Carbothermal reduction procedures at various temperatures from 600° to 900 °C were investigated. High temperature treatments caused, in most cases, the occurrence of a primitive cubic ordered ($L1_2$) phase in contrast to the disordered face-centered cubic (fcc) structure obtained at low temperature. The electrocatalysts were subjected to a pre-leaching procedure to modulate the surface properties. The catalyst based on 50% Pt_3Co_1/KB characterised by primitive cubic ordered structure showed superior characteristics both in terms of performance and stability. The PtCo alloy catalyst allowed a full achievement of the project milestone MS11 "First new catalyst materials demonstrating improved performance and/or stability provided to WP5 with Mass activity >2 fold improved over bench-mark catalysts and active surface area decrease in accelerated stress tests at least 50% lower than benchmark catalysts". The recorded mass activity was better than 0.46 A/mg at 0.9 V RHE at 80°C with 50% RH. This was better than the project target. It is notable that these milestones were achieved with practical catalyst loadings in the electrodes of 0.2 mg cm^{-2} useful to reach the MEA performance milestones.

Another milestone was dealing with "Performance loss less than 5% after 10,000 electrochemical cycles". No detectable performance degradation within the error limit (<1%) was observed using the primitive cubic order structure 50%/ Pt_3Co_1/KB cathode after 10^4 cycles at 80°C between 0.6V-0.9 V

RHE. A potential of 0.75 Volt was reached at 1.5A/cm^2 for the PtCo8T/KB catalyst achieving an ORR overpotential <0.43V (IR-free) at 1.5A/cm^2.

In conclusion, the main objectives for the anode and cathode development in this work package have been achieved. The electrochemical data set, showed improved performance for CNR catalysts with respect to the benchmark catalyst. Moreover, this improvement was also achieved by an optimisation of electrode structure allowing a better integration and an enhanced compatibility of the new developed materials such as electrocatalysts, membranes and ionomer.

WP5: MEA Development and Fabrication

The objective of Work Package 5 was to develop the understanding of MEA fabrication, particularly aiming to improve MEA performance and durability when targeting low platinum group metal (PGM) loadings. This was undertaken by incorporating the learnings and output materials from WP3 'Improvement and Development of materials: Polymers and GDL' and WP4 'Improvement and Development of materials: Catalysts' and producing fully integrated MEAs for cell testing. All MEAs were produced via the catalyst coated membrane (CCM) manufacturing method and were then delivered as developmental MEAs for single cell testing within the context of WP6 'Determination of Degradation Rates and Mechanisms' which informed the MEA development. Finally, the final stack MEAs were manufactured and delivered for the project defined stack testing within WP 8 'Demonstration of Stack Durability'.

Within this MEA development and fabrication work package, many aspects of MEA performance and durability were investigated. These included:

- Kinetic performance (low current density performance) and the trends as Pt cathode loading was lowered.
- Performance under mass transport limited regimes (high current density performance) and trends as Pt loading was lowered.
- Carbon support corrosion.
- Cell reversal tolerance (CRT).
- Tolerance to fuel impurities (low level CO tolerance).
- Improving membrane conductivity.
- Improving ionomer conductivity within the catalyst layer.
- Increasing ionomer stability within the catalyst layer.

One of the key results from this work package was to establish the degree to which the MEA could be thrifted to lower loadings without causing significant reduction in performance and durability. Reducing cathode loading from 0.4 mg cm^{-2} to 0.2 mg cm^{-2} showed a reduction in performance in line with the theoretical loss based simply on this loading reduction. The kinetic loss in moving from 0.4 mg cm^{-2} to 0.1 mg cm^{-2} was, however, not as great as theory suggests and it was assumed that this was a result of a high utilisation of the electrocatalyst in the thinner, lower loaded layers. The higher current density performance, however, did prove more problematic. It was seen within a systematic rainbow stack test that the anode loading could be thrifted significantly without any decay in performance, however, a reduction in cathode loading below 0.2 mg cm^{-2} showed significantly greater losses at high current densities. This is clearly demonstrated in Figure 8.
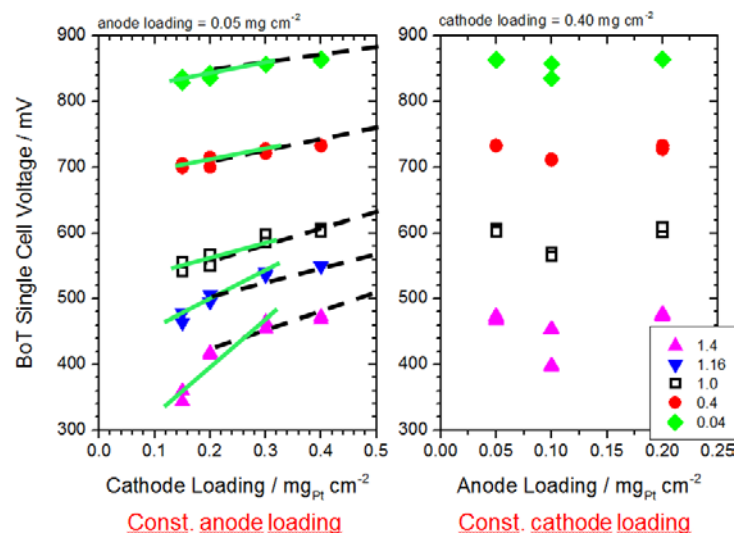
Figure 8. Increased mass transport encountered at high currents when moving to low cathode loadings, causing a greater than predicted loss in performance. In the panel single cell voltages versus Pt loading are plotted. The different data points in the left correspond to voltage values recorded at the indicated current densities (A cm^{-2}).

Another significant advancement made in WP5 was an understanding of the balance between improved performance and stability when selecting the properties of the ionomer to be included in the catalyst layer, in particular the equivalent weight (EW) of the ionomer. The nature of the ionomer included in the catalyst layers for proton transport can have an important influence on the MEA performance, not just by providing proton conductivity but also the ionomer film structure can affect gas movement and water handling properties of the layer. Therefore, it was important to develop the optimum layer structure to ensure optimal performance of the MEA. In addition, assessment of the stability of the layer structures was important to avoid excessive degradation of MEA performance over time.
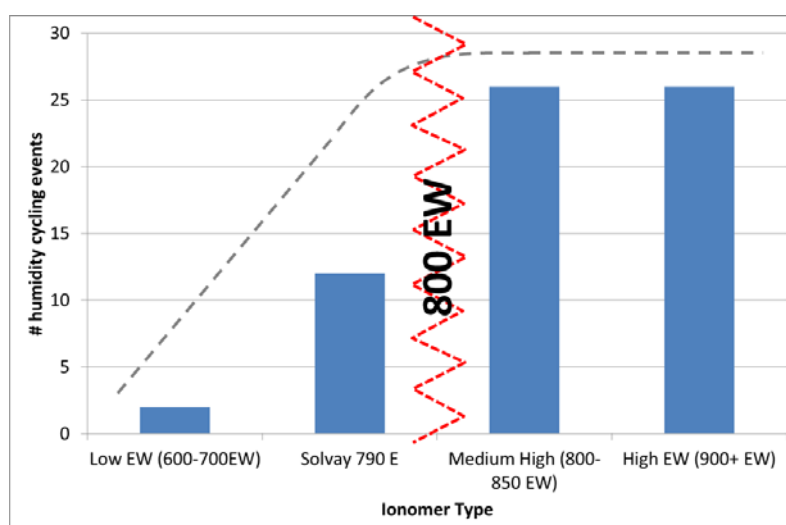

Figure 9. Trend in durability with increasing ionomer EW in catalyst layer clearly demonstrating that ionomers above 800 EW show increased stability.

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To improve cell performance, a transition to employing lower EW ionomers in the membrane was made. This was undertaken with a view to incorporating more acid sites and hence a higher iononic conductivity material. It was observed, however, that the lower EW ionomers gave significantly higher MEA degradation rates. When a systematic study into degradation rate and ionomer EW was carrier out there appeared to be a clear trend of stability with ionomer EW where the stability of the ionomer increased with increase ionomer EW until ~800EW, at which point there was a plateau in ionomer stability. The trend is clear in Figure 9.

Both these results informed the design of the final stack MEAs where a 0.2 mg_{Pt} cm^{-2} loaded cathode was incorporated with 830EW ionomer catalyst layer, as it was believed that this combination would provide the best balance of performance and durability at low loadings.

Throughout the project a number of developmental MEAs were manufactured in WP5 and supplied into the WP6 testing. Within this testing a summary of the performance of all project MEAs was collated. It can be seen from Figure 10 that developmental MEAs 4b and MEA 5 gave a performance very similar to that of the non-automotive commercial reference MEA, MEA0467 , which had a much higher cathode Pt loading of 0.4 mg cm^{-2}. The performance achievement of the final project MEA 5, giving similar performance to the commercial reference at high currents, was due to the realisation of the conductivity benefit from the inclusion of the thinner 10 µm membrane, as well as the significant optimisation of the cathode catalyst layer.
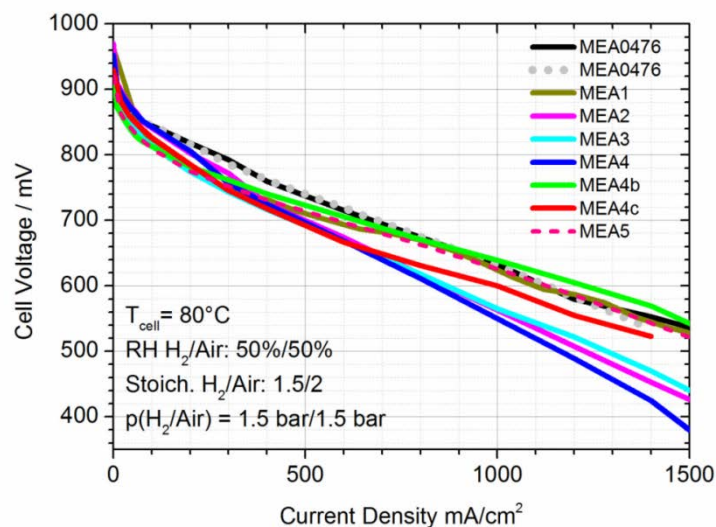
Figure 10: Polarisation performance of principal developmental MEA types supplied by JMFC and tested under IMPACT defined test conditions in single.

In a specific test conducted at GIST (Figure 11), the performance of the lowest loaded MEA4c was measured up to a current density of 2.5 A cm^{-2}. This shows that the power density target of 1.0 W cm^{-2} was achieved at 1.8 A cm^{-2}.

Figure 11: Performance of MEA4c measured at 80°C cell temperature, 50%RH, 1.5 bar absolute pressure and H$_2$ and air stoichiometries of 2.0.

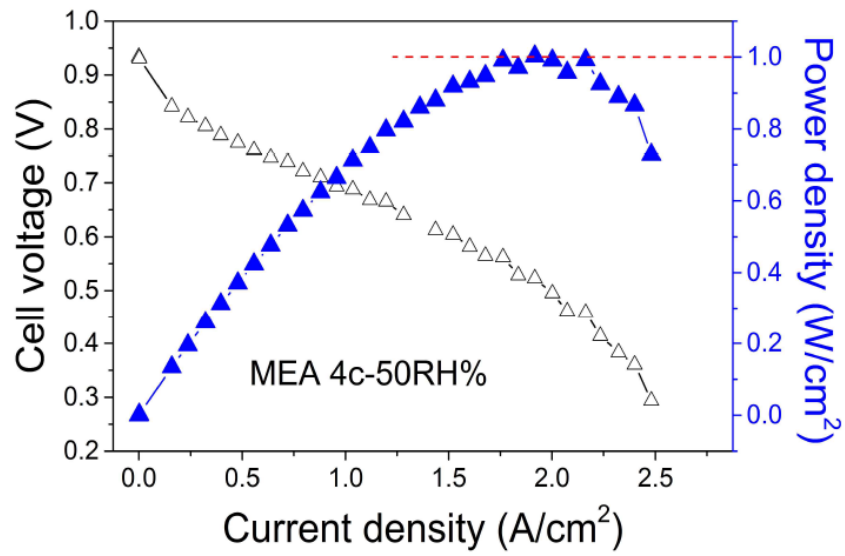## WP6: Determination of Degradation Rates and Mechanisms

The aim of WP6 was the precise determination of degradation rates and durability improvements of MEAs operated in automotive conditions as well as better understanding of reversible and non-reversible degradation mechanisms. A further goal was to propose strategies to mitigate degradation and to recommend most durable MEAs developed in WP5.

The durability tests of MEAs have been conducted using the dynamic FC-DLC protocol in single cell configuration for 500 – 1000 h. A typical test consists of several test blocks of continuous FC-DLC operation (see WP8) followed by a refresh procedure to recover reversible performance losses as sketched in Figure 12.
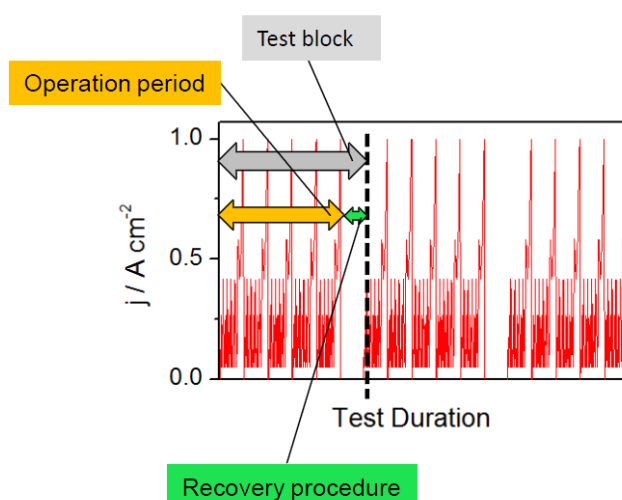

Figure 12: Scheme of the electrical load profile of a typical dynamic durability test consisting of numerous test blocks of continuous FC-DLC operation followed by a refresh procedure.

Thanks to the regular refresh procedures it is possible to discriminate between reversible and irreversible degradation as demonstrated in Figure 13. Thereby, the latter is the one primarily limiting the MEA lifetime for automotive applications. Hence the irreversible degradation has to be taken into account when assessing durability improvements making the regular refresh a necessary part of each durability test. A dedicated study was undertaken to investigate the reasons for the recovery of reversible voltage losses. According to the current knowledge, the main contribution to the recovery is due to soaking of the catalyst layer in water and due to removal of chemisorbed contaminations from the catalyst.

The irreversible degradation rates of selected IMPACT MEAs are provided in Figure 14. All new MEA variants demonstrated durability improvement compared to the commercial reference MEA. The very promising stability of MEA4c led to its selection for the final stack testing as MEA 5 with slightly increased Pt loading to further improve its stability, in Work Package 8.
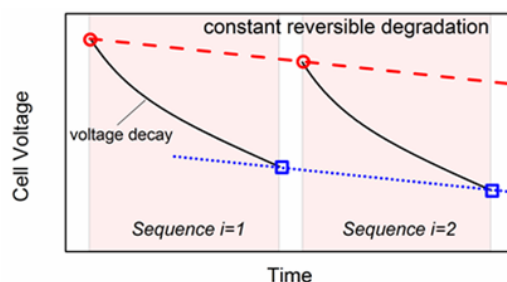
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Figure 13: Voltage decay during two subsequent sequences (test blocks) interrupted by a refresh procedure. Reversible degradation is represented by the back curve that occurs during continuous operation. Irreversible degradation corresponds to the linear regression of voltage values before or after refresh (red or blue lines).
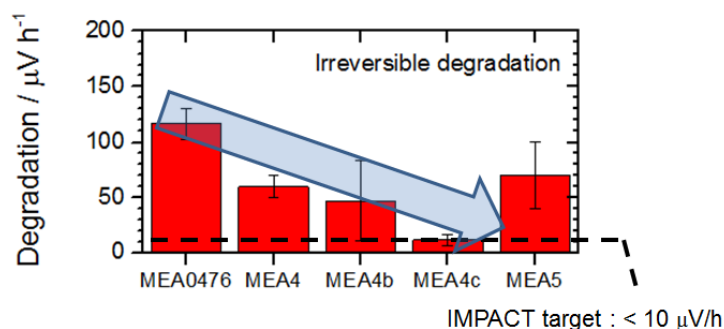

Figure 14: Irreversible degradation rates of IMPACT low Pt loaded MEAs (generations 4-7) compared to commercial reference MEA (non-automotive) determined at 1 A cm^{-2} in single cell configuration.

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Further work performed in the frame of WP6 was a study about the relation of degradation and water management as well as the influence of impurities. To visualize the local current density distribution during operation at different humidity levels DLR's segmented cell was used.

A very important aspect is the effect of catalyst loading on durability and performance which has been studied in a rainbow stack equipped with differently Pt loaded MEAs. A plot summarizing the irreversible degradation rates at different Pt-loadings and different current densities is provided in Figure 15. As expected, the highest degradation is observed at high current densities and low loadings. Thereby, a clear and sharp threshold between high and low degradation rates occurs at 0.2 mg$_{Pt}$ cm^{-2}. Below this cathodic Pt-loading a sudden increase of the irreversible degradation rate is measured especially at current densities approaching 1 A/cm^2.

Since most MEA tests are performed in single cell configuration for cost reason, but eventually the MEAs will be operated in stacks, it is of importance to assess the comparability of single cell and stack measurements regarding durability and performance. Especially the durability depends very much on various factors that do not depend on the MEA itself such as cell design, applied cell compression (and its possible relaxation during operation, …). Hence, a careful analysis is needed to identify external influences contributing to measured degradation rates of MEAs. In this context, a public preliminary report on comparability of single cell and stack test has been prepared.
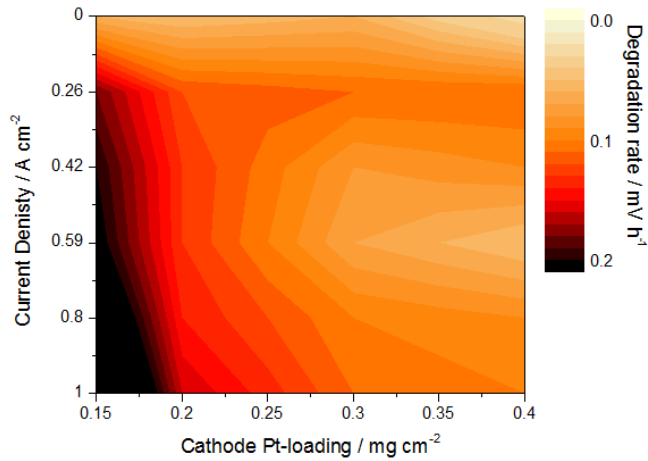
Figure 15: Pseudo 3D plot showing the irreversible degradation rate (color scale) versus current density and cathodic Pt-loading. The degradation values at loadings 0.15, 0.20, 0.30, 0.4 mg_{Pt} cm^{-2} are real experimental data; the values at loadings 0.25 and 0.35 mg_{Pt} cm^{-2} are interpolated data.

In order to understand the reasons for MEA degradation, ex-situ analysis has been performed on new and aged samples. In the following several interesting findings are outlined.

In the case of membranes, for instance, areas with low stiffness were detected indicating ionomer degradation, often neighboured to Pt particles/agglomerates stemming from the electrode and redeposited in the membrane.

Regarding electrode degradation ionomer layer thickness was analysed; after operation, it exhibited a significant thinning especially for the first generation MEA. Thereby, the decrease of ionomer layer thickness was more severe at the anode than at the cathode side (Figure 16). Correlations of the initial ionomer thickness and initial performance with total and irreversible degradation rates of the MEAs were found. The investigation of ultra-thin model layers in the electrodes gave hints for differences in structure and conductivity with thickness.
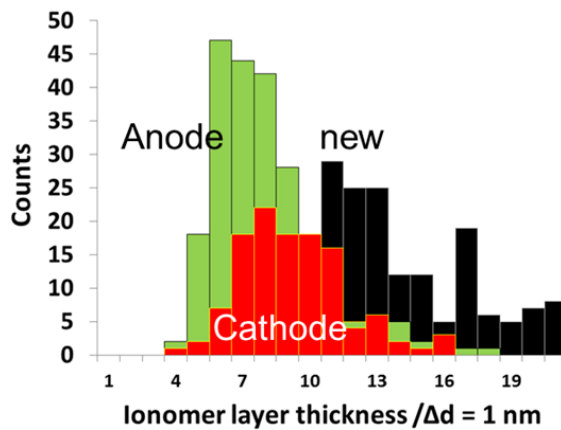

Figure 16: Ionomer layer thickness of new and degraded anode and cathode of project MEA1 after 235 h of operation.

WP7: Modelling and Mitigation Strategies

An innovative model of a cathode PEMFC has been proposed, coupling a pore network model (PNM) for describing the two-phase transfers in the diffusion medium (DM) of a GDL to a continuum approach in the MPL, catalyst layer and membrane. The advantage of the PNM used here is to allow the modelling of the liquid water formation by condensation in the DM using a state-of–the art condensation algorithm.

The present study has allowed confirming that more liquid water is located close to the rib/GDL interface due to condensation. In addition results show that it is important to take into account the coupling with the CCL as the current density profile at the CCL/GDL interface is essential for simulating the performance of the MEA. This is true when no liquid water is inside the GDL (dry condition) for high current densities, but this is crucial in wetter condition when liquid water appears in the GDL.

Another result is that, as suspected, the liquid water pattern can be a first order parameter for the performance of the MEA. Consequently, as the two-phase patterns are fully different between the condensation scenario in the GDL (more liquid water is close to the rib/GDL interface than to the CCL/GDL interface) and the injection scenario (more liquid water is close to the CCL/GDL interface than to the rib/GDL interface), this confirms that the condensation and evaporation phenomena need to be taken into account. These results are consistent with some experiments (Figure 17).
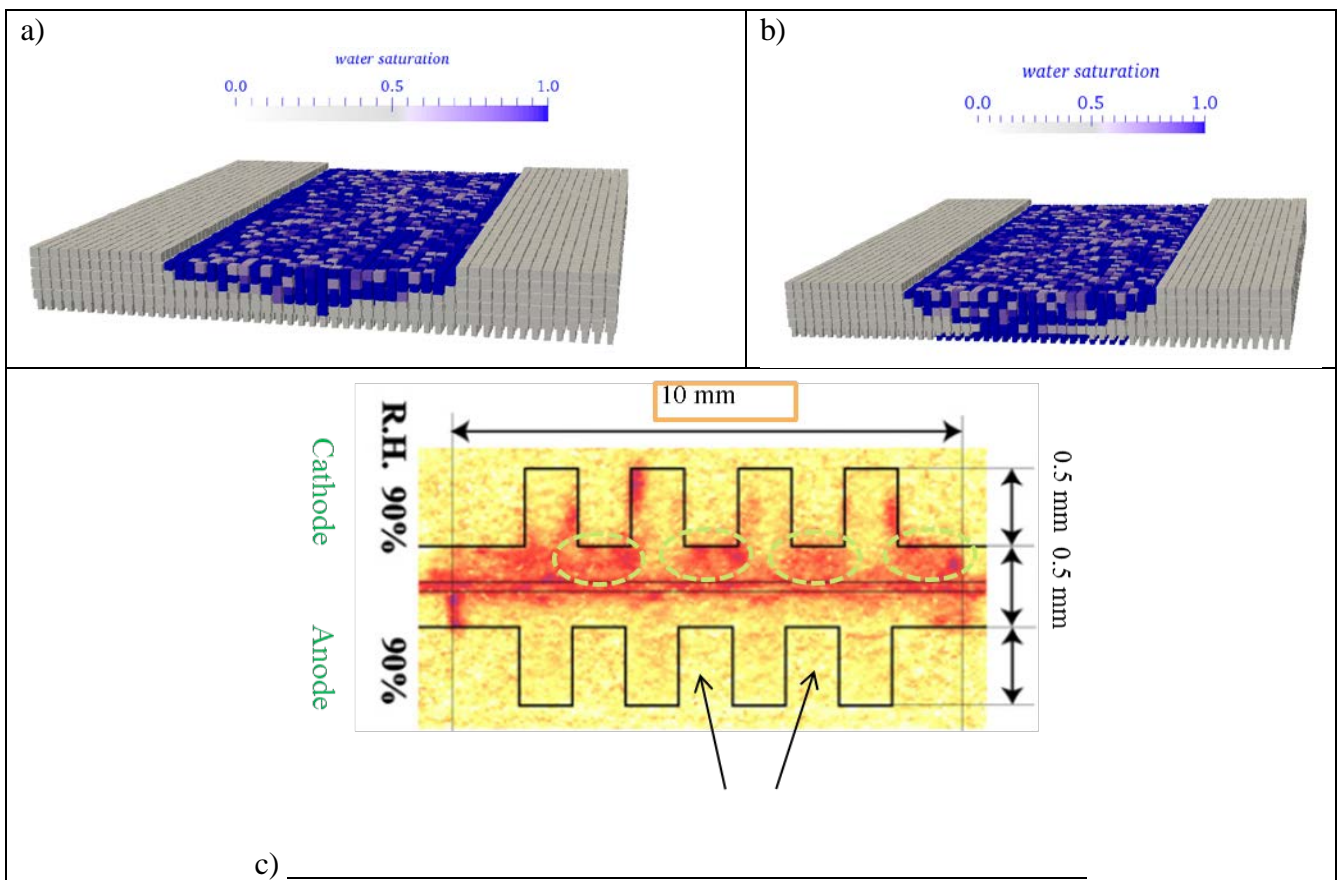

Figure 17 : Liquid saturation distribution under wet condition (liquid water is in blue) for the a) coupled model, b) non-coupled model, and c) experiments[3]

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[3] Boillat, EC 2008

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A mitigation strategy has been analysed by introducing Pt dissolution look-up tables into cell model and defining optimal trajectories for the operating conditions of a stack. Results show that tuned operating parameters (RH, pressure) can help improving durability of a PEMFC.

To amend results with a direct relation of the porous material structure to appearance and behaviour of liquid water within the material and to further investigate ageing effects, an advanced Monte-Carlo model has was employed and further developed in the project. This model relies on material structure data as obtained from state-of-the art imaging techniques as X-ray tomography or FIB-SEM for the nano-scale to simulate the liquid water distribution within the material. In doing so, the model takes into account evaporation and condensation effects on the basis of the locally resolved thermodynamic boundary conditions (temperature, relative humidity, pressure). These were obtained from sophisticated computational fluid dynamics modelling of the full cell, which provides the coupling of the different layers on the different length scales in this case. During the project, the MC model was significantly improved, the most valuable new feature being the possibility to perform simulations on variable length scales ranging from the µm-scale relevant for the GDL substrate down to the tens of nanometres scale applying to the relevant pores of the MPL and the catalyst layer (cf. Figure 18).
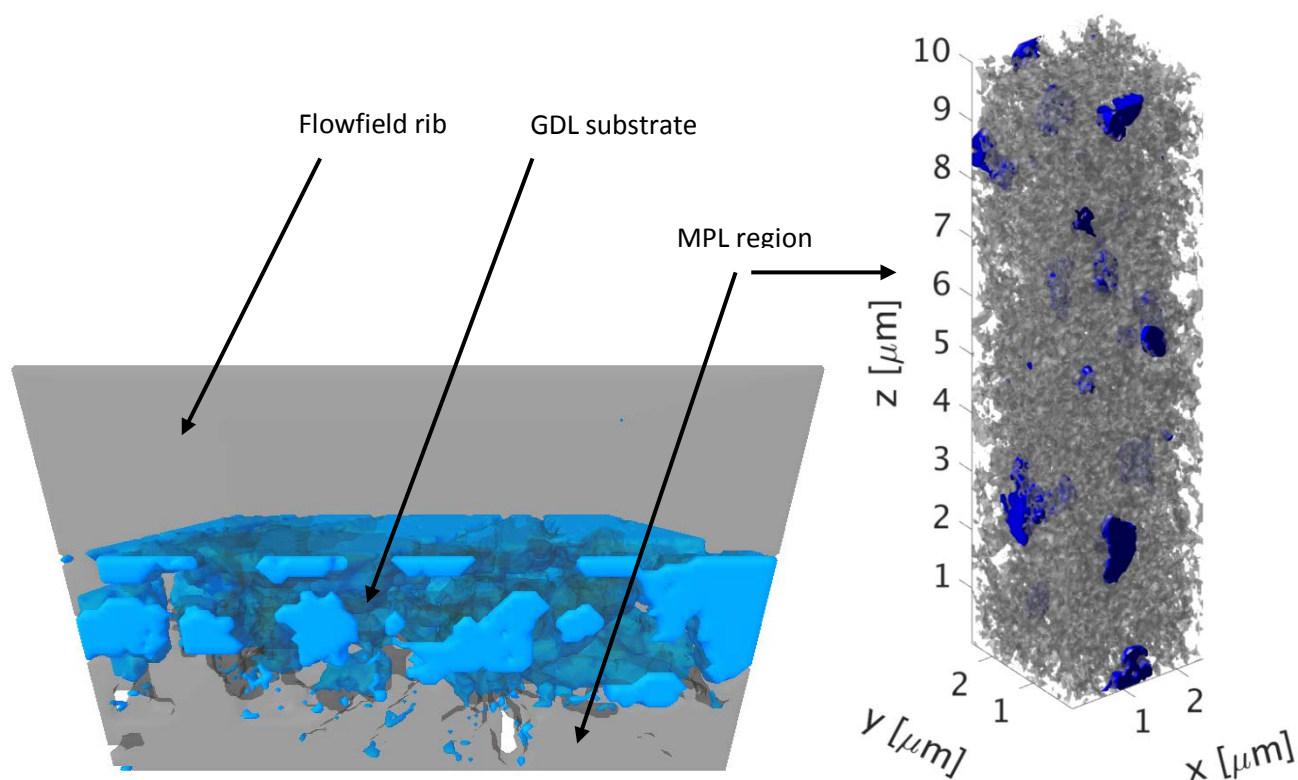
Figure 18: Monte-Carlo (MC) modelling results on different length scales: resulting water distribution in several ten µm-scale pores in the GDL substrate below the rib (99.5%RH) (left) and in several ten nm-scale pores in the MPL (80% RH).

Comparing the results obtained for the GDL substrate to those for the MPL, it can be said that considering water transport from the catalyst layer into the MPL in the gas phase, condensation within the MPL becomes relevant at lower RH values than for the GDL substrate, probably this is due to capillary condensation effects. Nevertheless, even going to high RH values near 100%, no

complete filling of the structure occurs and still some open space for gas diffusion remains. Even though water transport into the structure is considered only in the gas phase and the overall filling degree for this condensation scenario does not significantly depend on the wettability of the material, the results obtained for the MPL show that for hydrophilic contact angles, a completely different distribution of the liquid phase, which is formed by condensation, occurs. Though the capillary condensation first appears in the very small pores, the bigger ones are preferably filled at higher RH in the hydrophobic case. In contrast, for hydrophilic surface properties, the smaller pores are the ones which are nearly completely filled. Thus, to maintain operability even after many operating hours, first of all the hydrophobic properties of the material should be preserved as long as possible. But furthermore, it can be concluded that a mixed, i.e. broad pore radius distribution would be beneficial for the MPL. This study suggests that larger pores would ensure free pore space for gas diffusion under non-condensing conditions when for aged material, the hydrophobicity of the material may be partially lost. On the other hand, small pores are important in the case of fresh (i.e. hydrophobic) material, as they can be expected to remain open under high RH conditions.
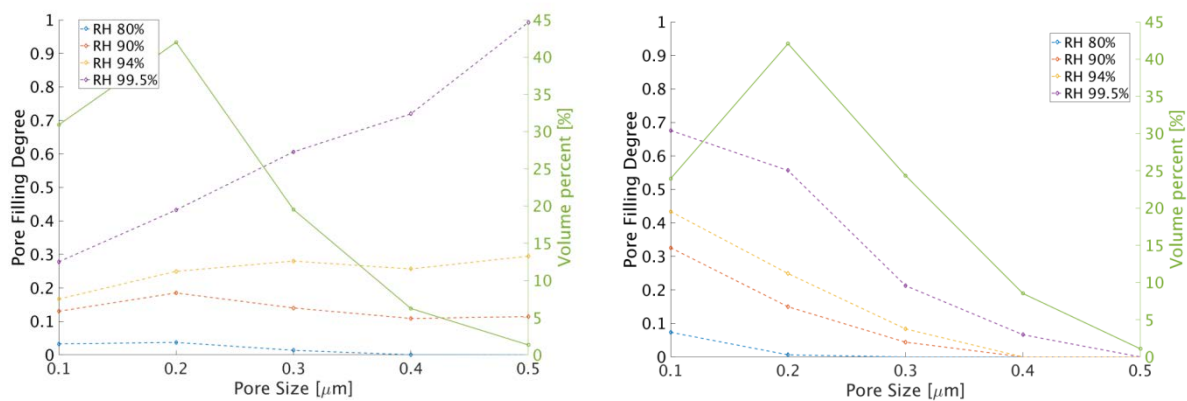
Figure 19: MC results on the relation of pore filling degree to pore size at different humidity levels for the MPL. Left: fresh material (hydrophobic, contact angle: 94°), right: aged material (hydrophilic, contact angle: 80°).

Thus, the information related to the detailed inner pore structure which was obtained by MC modelling allowed proposing mitigation strategies rather related to material development, whereas the coupling of length scales as achieved for the innovative PNM model permitted developing mitigation strategies related to the stack operating conditions.

WP8: Demonstration of stack durability

The main objective WP8 was to perform the endurance test on the short stack equipped with Reference MEAs and IMPACT MEAs. Testing of advanced material and on the impact of certain stressors was evaluated in the second period of the project.

- The first activities in WP9 were the adaptation of the designs and manufacturing of the stacks necessary in order to perform durability testing of reference and project MEAs. Secondly, a dynamic Round Robin test based on the FC-DLC protocol has been conducted using a ZSW stack according to the definition of the detailed testing procedure including duration and relative load of all load points which can be find in the public deliverable D6.4. The overall result of the Round-Robin Test performed at constant stoichiometry and constant flow rates (see figure) is presented in Figure 20. The irreversible degradation rates are evaluated in the three load levels 5, 42 and 100% with 100% corresponding to 1 A/cm^2. From the test following conclusion can be drawn: Constant flow mode during dynamic load cycling was the dominating stress factor and not the dynamic load cycle itself.

- Regular shut-down times for stack recovery after performed test blocks in constant-flow mode were not sufficient for full recovery of reversible losses.4

- For the evaluation of irreversible degradation during the Round-Robin-Test, values from polarization curves were more reliable; supposed to be significantly less affected by test equipment, differences in test operation scripts or switches of test benches.
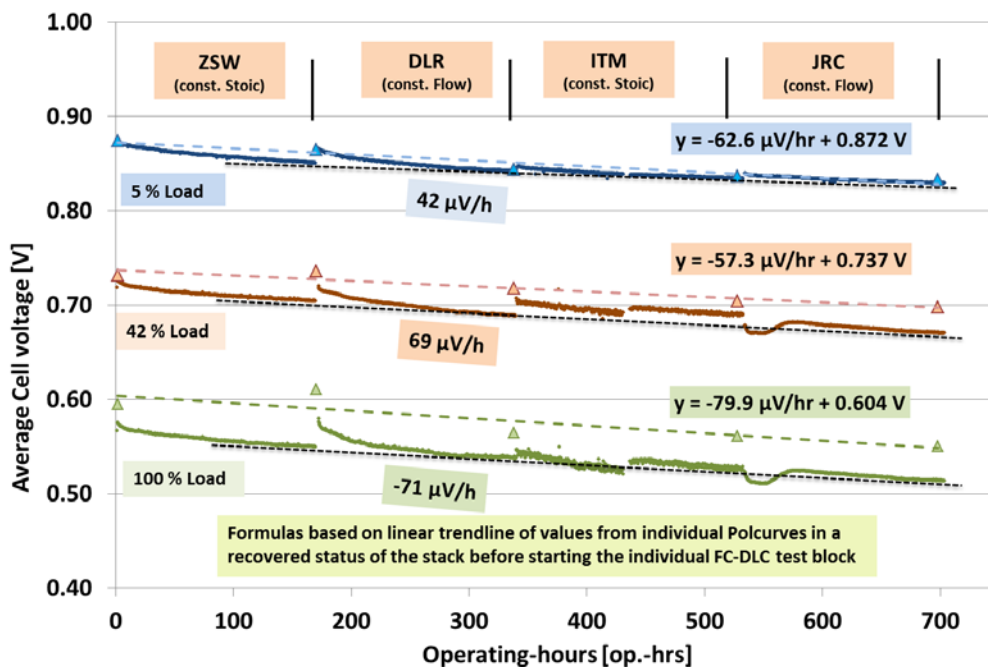

Figure 20: Result of the Round-Robin Test campaign

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[4] Back home at ZSW after the Round-Robin-Test, only after the "End Of Test"- characterization during regular FC-DLC-operation in constant-stoic-mode a "new" recovered and stable cell voltage level was found, staying stable over the whole subsequent performed durability test of 2.500 hrs.

Subsequently, several of the MEAs developed within the project were tested at stack level to obtain comparable performance and degradation data. Though the ultimate performance target of 675 mV @ 1.5 A/cm² was neither reached by the project MEAs, nor by the commercial reference MEA, MEA IVc showed a clearly better performance at BOL than the Reference MEA, even though its Pt loading of 0.2 mg Pt/cm² was really low and thus meets the project targets. Also MEA V with a slightly higher Pt loading of 0.25 mg Pt/cm² showed a better performance at BOL than the commercial reference MEA. Unfortunately, still both the considerably higher reversible degradation within the test blocks and the higher irreversible degradation rates constitute significant drawbacks of the low loaded MEAs. Consequently, they failed to reach the durability targets of the project. Specifically, the test had to be stopped after 1700 FC-DLC operating-hours due to instability of some cell voltages at the 100% load-point. Figure 21 directly compares the durability test result of the project MEA V and the commercial reference MEA. Clearly, the project MEA V suffered from unexpectedly higher degradation than was anticipated and the end-of-life was reached considerably earlier than the intended durability of 5000 h. Unfortunately, there was no time left at the end of the project to investigate the exact reasons for the unexpected behaviour. The latter is also true for the reference MEA which sustained at least for 2500 h, but the lower loaded MEA failed still earlier.

Still, further research activities should be conducted to investigate the significance of the test procedures employed here for real vehicle operation. Possibly the relatively long uninterrupted operation and the FC-DLC test cycle employed here impose stress factors that lead to accelerated ageing especially in the case of low loaded MEAs and the 'real life' durability of the latter may have been somewhat underestimated.
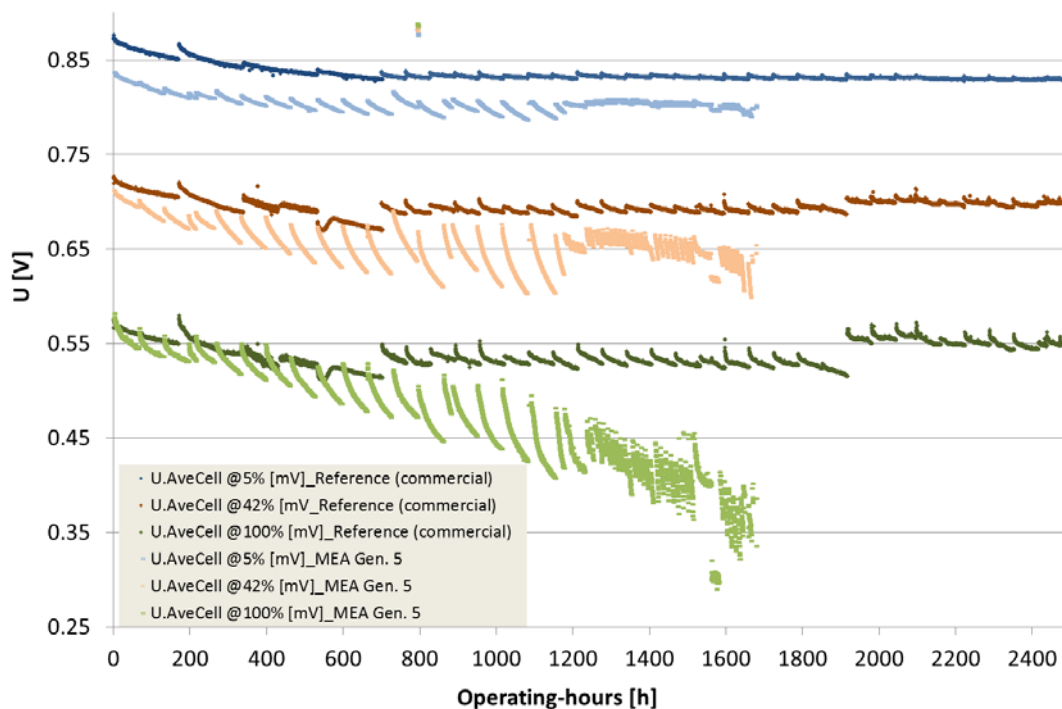

Figure 21: Result of the durability-test for MEA V, compared to the commercial reference MEA.

Potential Impact and main Dissemination and Exploitation Activities

Results of the project have been published in the scientific literature (15 papers) and presented in scientific conferences (36 contributions) as listed in Section 4.2 A.

Two workshops were organized in the course of Impact – one in Toulouse in 02/2015 which was associated with the FDFC conference and a second one in Stuttgart in 10/2016 associated to the congress "World of Energy Solutions". The presentations were covering a broad range of topics including modelling, materials synthesis, analytical techniques and cost analysis.

Moreover, a summer school was organized in Grenoble on September 22nd and 23rd. It was attended by 24 participants, 10 of them were students. A total of 11 lectures covered a broad range of topics related to lifetime issues of PEM fuel cells. In addition, the students attending the summer school reported on their current work.

The following table intends to give a summary of some lead questions which have been addressed within the IMPACT project as well as suggestions for further research.

Topic	Status of Understanding after IMPACT	Open Questions
Reversible degradation	In all durability tests reversible degradation has been observed as non-linear voltage decay at a given load with time. The shape of the reversible degradation curve changes with time indicating that the processes responsible for reversible voltage loss are also changing with time. This can be seen in the evolution of the parameters of an empirical degradation model. Reversible degradation can be caused by water management issues, oxidation of the cathode catalyst surface, contamination of the anode catalyst surface, some fuel or air contaminant etc. There are also indications for re-distribution of the ionomer in the electrode. In order to reverse the effects of reversible degradation, all these aspects need to be addressed in a recovery procedure. Endurance testing and separation of irreversible and reversible degradation phenomena eventually allowing to assign proper degradation rates to a given MEA still poses a challenge. The test protocol used as well as the operating conditions do have a significant influence on the degradation rates observed. More insight is needed on the impact and interaction of test procedures and operating conditions.	Despite progress made in this project, the individual mechanisms leading to reversible degradation are not fully understood. Although empirical modelling provided some insight, the meaning of the empirical parameters as well as their interaction with irreversible degradation phenomena need further investigation. The results of endurance testing are strongly depending on the testing protocol used. This includes precise control over the load free phases. Still, further research activities should be conducted to investigate the significance of the test procedures employed here (FC-DLC) for real vehicle operation. Possibly the relatively long uninterrupted operation and the FC-DLC test cycle employed here impose stress factors that lead to accelerated ageing especially in the case of low loaded MEAs and the 'real life' durability of the latter may be underestimated.
Irreversible degradation	Irreversible degradation can be linked to catalyst surface loss caused by particle growth, catalyst corrosion and substrate corrosion as well as loss of sulfonic groups causing membrane degradation. Fuel and air contaminants can also contribute, yet such effects might be slowly reversible. Irreversible degradation also appears to be non-	A better understanding of the impact of individual degradation phenomena on irreversible degradation is required. Furthermore, a better link to performance modelling would be necessary. Further work to differentiate reversible from irreversible

Topic	Status of Understanding after IMPACT	Open Questions
	linear with time. The amount of ionomer available in the catalyst layer positively influences the begin of life performance, yet enhances degradation. Surprisingly, degradation of the ionomer in the catalyst layer seems to have a large influence on overall yet little effect on irreversible degradation. Imaging showed changes at the catalyst layer / MPL interface in an aged sample indicating changes in water management and mass transport.	degradation will also require the development of test procedures beyond the level established in IMPACT and previous projects. Better understanding of the the impact of GDL aging to improve methods of mitigation are needed. This includes a correlation of microstructure and surface properties evolution.
Analysis / modelling of degradation kinetics	Degradation kinetics could be successfully modelled via an empirical model coupling linear and exponential processes. The parameters describing the model show a clear evolution with time. Pore network modelling gave insight into catalyst corrosion. The importance of taking condensation phenomena into account could be established. The importance of surface properties could be established as well. Imaging methods were found to provide geometric data for the MPL and the catalyst layer down to the nanometer scale.	Further progress in modelling methods is required to understand the interaction of different phenomena observed to be relevant for degradation. Furthermore, analytical or modelling tools are required to determine localized surface properties in the length scale the models are applied.
Role of electrode composition Catalyst	According to the Kinoshita theory, the optimum Pt catalyst particle size for the oxygen reduction reaction is between 2.5 and 3.0 nm. The motivation is that being the oxygen reduction a structure sensitive reaction, the presence of a large concentration of defects in very small particle decreases the specific activity whereas a poor dispersion (large particles) decreases significantly the mass activity. On the other hand, slightly larger Pt particles generally obtained in high temperature annealing treatments of alloys can reduce catalyst degradation as observed for PtCo catalyst with Pt segregation on the surface. However, at high potentials e.g. 1.2 V the role of the carbon support in determining the extent of degradation is predominant. Use of graphitic carbons can reduce substantially carbon corrosion. When moving to platinum loadings of less than $0.2 \ \mathrm{mg \cdot cm^{-2}}$, additional mass transport effects have been observed.	Use of highly conductive oxide supports may strongly reduce support corrosion and thus catalyst degradation at high potentials. Open questions regard electronic conductivity and dispersion of the active phase as well as surface interaction of the support material. Better understand additional mass transport effects encountered when moving to ultra-low catalyst loadings of less than $0.2 \ \mathrm{mg \cdot cm^{-2}}$.
Role of electrode composition Electrode treatment	Coating of the catalyst layer with oxides by ALD or grafting by a hydrophobic agent also improved the corrosion resistance of the catalyst layer.	Electrode additives or coatings protecting the support surface from electrolytic contact should be investigated more closely.

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Topic	Status of Understanding after IMPACT	Open Questions
and additives		
Extent and impact of ionomer degradation inside the electrode	Clear indications of the influence of ionomer equivalent weight, content and mobility in the electrode layer on performance and degradation could be established by AFM. Furthermore, differences in the ionomer nanostructure could be observed indicating impact on electrode performance.	The methods to study ionomer distribution in the catalytic layer as well as the structure and dynamics of ion conduction in the catalyst layer need further investigation.
Correlation of ECSA with MEA at high current density	ECSA is mainly affecting the mass activity in the activation controlled region i.e. the region at low current density. Catalyst properties are essentially affecting the large potential loss in this region. However, the properties of the catalyst also reflect on the other regions even if in a minor extent. In particular, at high current density a high catalyst dispersion may help oxygen access to the catalytic sites and increase the turn over frequency.	Depending on the method ECSA is determined, also catalyst surface buried under a thick ionomer layer can show electrochemical activity. A direct relationship is not yet available since the performance at high currents depends on several phenomena and deconvolution of all these contributions in a precise way will need further modelling efforts as well as experimental validation.
Correlation of catalytic activity measured ex-situ with MEA-performance at high current density	Electro-catalyst formulations screening in rotating disc electrode is a fast and useful tool for a preliminary assessment of catalyst activity and stability. However, the observed catalytic activity cannot be used to provide a precise estimation of the catalyst performance in fuel cell. The reasons are that the catalyst loading in RDE is not exactly determined as it is in a gas diffusion electrode or in a catalyst-coated membrane procedure. Phenomena like non homogeneous catalyst slurries with the ionomer, poor transfer to the RDE tip, application of additional ionomer films can strongly affect the results in RDE. In this regard, mass activity for oxygen reduction at the cathode (rate determining step) can be properly determined at the single cell level. The procedure involves use of hydrogen at the counter and reference electrode. Moreover a correction of the polarization curves for hydrogen cross-over and ohmic resistance is necessary. Thus, the mass activity is derived by plotting polarization curves as IR and cross-over –free Tafel plots. This information is obtained in situ and can provide clear assessment of the catalyst performance for practical application. Determination of ECSA in situ by cyclic voltammetry can also provide information about specific activity. On the contrary, the performance recorded for the RDE at high currents in a flooded configuration cannot be compared or transferred to that observed using a gas-diffusion electrode where the transport control regime and gas solubility at the interface are	Activation of the electrode in-situ, by cyclic voltammetry, can, in the case of Pt alloys, remove non noble metals segregated on the outer catalyst surface thus increasing the surface roughness with a performance that may be different from that achieved in a stack configuration that is not activated using similar procedures. Better understand the impact of mass transport effects in the catalyst layer, improve catalyst layer additives and treatments to improve the catalyst layer stability and the high current density performance of electrodes using alloy type catalysts.

Topic	Status of Understanding after IMPACT	Open Questions
	completely different.	
MPL/GDL-interaction	Morphological changes at the boundary of MPL and the catalyst layer have been observed in aged samples. Furthermore inhomogneities in Pt-distribution in the catalyst layer were detected.	A comparative assessment of improved imaging methods should be carried out. Ideally, a method sensitive to surface properties such as wettability in the relevant length scale should be developed to validate modelling results.
Evolution of catalyst layer and GDL surface properties / wettability	Imaging by FIB-SEM and AFM gave clear indications of changes in layer morphology for the catalyst layer and the MPL in aged samples.	Establish a correlation of ionomer / membrane swelling behaviour on endurance. Establish a correlation of GDL surface properties on performance and endurance.

Potential Impact and Exploitation by Solvay

Solvay Specialty Polymers S.p.a. (SLX) is one global business unit of Solvay S.A. and it is now mainly focused on different special materials such as: fluorinated polymers, aromatic polymers, high-barrier polymers and compounds.

Solvay as a leading worldwide chemical company strongly supports the sustainability and sustainable growth. The commitment of Solvay to fighting climate change was recently confirmed at COP 22 in Marrakesh where it was highlighted the important role that innovation from the chemical industry and collaboration between industry sectors can play to implement the Paris agreement of late 2015.

Solvay is also participating in a technology partnership with some top chemical companies to find new ways to reduce carbon emissions generated in their energy-intensive processes. This partnership aims at carrying out collaborative research and to evaluate the feasibility of different Carbon Capture and reUse (CCU) technologies.

Moreover, Solvay is going to cut the carbon intensity of its activities by 40% by 2025, with an intermediary target of minus 20% by 2018.

Solvay Specialty Polymers is very well aligned to the Company commitment to support the sustainability and sustainable growth with its material portfolio comprising products designed for, among the others, light weighting, green chemistry and sustainable mobility; fuel cells are commonly recognized as key player for the sustainable mobility in the next future and thus is very important that a sustainability-focused Company is present on the market, being involved in cutting-edge developments, collaborating and exchanging ideas with the main actors in this field.

Impact project is targeted at understanding the main mechanisms leading to MEA degradation, at improving its durability and at using at best the precious Pt catalyst, reducing, where possible, the loading. Cost and durability are very important issues that must be addressed before the massive

deployment of fuel cell cars and thus the results achieved within this project are highly valuable for the future exploitation.

In the frame of Impact, SLX has iteratively developed advanced Aquivion® PFSA-based membranes as a result of our efforts aimed at finding the best trade-off between electrochemical performance and durability. Furthermore, the last generation of membranes developed within Impact contain proprietary additives specifically designed to mitigate degradation and further increasing membrane lifetime.

As mentioned before, also ionomer dispersions have been improved in order to obtain the maximum advantage in terms of performance and stability within the electrodes.

Advanced ionomer dispersions, used for reinforced membrane manufacturing, can be surely commercialized opening new commercial lines, too.

The technical skills and know-how acquired during the project and used to develop, improve and prepare also at larger scale ionomer dispersions, reinforced membranes and additive are of capital importance for SXL and will be valuable to produce and make available on the market more performing and more durable materials for PEM fuel cells giving a significant commercial advantage for Solvay.

Potential Impact and Exploitation by Johnson Matthey

In the coming years, fuel cell component design will continue to evolve as fuel cell technology becomes a part of our everyday existence and Johnson Matthey Fuel Cells is committed to be at the forefront of this development activity. Whilst JMFC is committed to a broad range of fuel cell applications, the automotive sector remains the primary target of the business.

Fuel cells in transportation applications are coming of age. Repeat customers are purchasing systems for materials handling applications without government subsidies in North America, an increasing number of fuel cell buses are operating in cities across the world and, for the first time, fuel cell cars are rolling off series production lines.

The work targeted within IMPACT looked to understand and improve the durability of MEAs with low platinum concentrations at typical automotive operation conditions. The delivery of low loaded MEAs is essential for widespread commercialisation as platinum forms an important part of the cost of this component. The learnings from the IMPACT project have already informed the design of improved low loaded MEAs with enhanced lifetime stability. The exploitation of these new components and MEAs by JMFC at a commercial scale will involve a further 1-2 year phase of product/process development, and customer qualification.

JMFC has good working relationships with many of the OEMs currently establishing fuel cell electric vehicles into the automotive market. It is anticipated that OEMs will be interested in working with JMFC to evaluate and provide feedback on the performance of the novel improved stability MEA designs developed in IMPACT. They also have very detailed and carefully staged new product introduction cycles and qualification protocols. Thus JMFC will have excellent visibility of the commercial prospects for the new MEA designs and the timescales on which significant volumes of new products will be required. The eventual fabrication of new catalyst layer

and full MEA products will be established in the UK at JMFC's Swindon MEA manufacturing facility, and will have a positive knock-on effect to a wide range of suppliers and support service industries.

One specific example of the exploitation of new components developed in the project is the developments in ionomer stability which has already informed the MEA design of pre-commercial products currently being tested at JMFC. This learning has directly affected the development of future commercial MEA products which will mean the ideal balance between performance and durability can be struck and hence provides a significant commercial advantage. In addition, the learnings on the limit to which the cathode Pt loadings can be thrifted with the current generation of catalyst layer designs, without encountering large performance losses at high current densities, will also inform the design of commercial MEAs in the short-term and help establish development priorities for future MEA developments.

Address of the Project Public Website and Relevant Contact Details

Project Public Website

A brief description of project objectives, structure and further information can be found on the project website: **www.eu-project-impact.eu/**

Relevant Contact Details

No.	Contact	Partner Logo	Country
1	*Project Coordinator:* Prof. Dr. K. Andreas Friedrich (andreas.friedrich@dlr.de) Deutsches Zentrum für Luft- und Raumfahrt e.V. (DLR)		Germany
2	Dr. Joel Pauchet Commissariat à l'Energie Atomique (CEA)		France
3	Dr. Georgios Tsotridis Joint Research Centre, Institute for Energy and Transport (JRC-IET)		The Netherlands
4	Dr. Anotnino S. Arico Consiglio Nazionale delle Ricerche (CNR)		Italy
5	Dr. John Blake Johnson Matthey Fuel Cells Limited (JMFC)		United Kingdom
6	Dr. Ludwig Jörissen Zentrum für Sonnenenergie- und Wasserstoff-Forschung Baden-Württemberg (ZSW)		Germany
7	Prof. Dr. Renate Hiesgen University of Applied Science Esslingen (UAES)		Germany
8	Dr. Henning Markötter TU Berlin (TUB)		Germany
9	Dr. Marc Prat Institut National Polytechnique de Toulouse (INPT)		France
10	Prof. Dr. Jaeyoung Lee Gwangju Institute of Science and Technology (GIST)		South Korea
11	Dr. Claudio Oldani Solvay Specialty Polymers Italy S.p.A (SLX).		Italy

4.2   Use and dissemination of foreground

Section A (public)

This section includes a list of all scientific (peer reviewed) publications relating to the foreground of the project (A1) and a list of all dissemination activities (publications, conferences, workshops, web sites/applications, press releases, flyers, articles published in the popular press, videos, media briefings, presentations, exhibitions, thesis, interviews, films, TV clips, posters), see A2 below.

	A1: LIST OF SCIENTIFIC (PEER REVIEWED) PUBLICATIONS, STARTING WITH THE MOST IMPORTANT ONES									
NO.	Title	Main author	Title of the periodical or the series	Number, date or frequency	Publisher	Place of publication	Year of publication	Relevant pages	Permanent identifiers[5] (if available)	Is/Will open access[6] provided to this publication?
1	*Quantitative in Situ Analysis of Ionomer Structure in Fuel Cell Catalytic Layers*	*T.Morawietz*	*ACS Applied Materials and Interfaces*	*Volume 8, Issue 40*	*American Chemical Society*	*USA*	*2016*	*27044-27054*	*DOI: 10.1021/acsami.6b07188*	*no*
2	*Structure and conductivity of fuel cell membranes and catalytic layers investigated by AFM*	*R. Hiesgen*	*Materials Research Society Symposium Proceedings*	*Volume 1774*	*Materials Research Society*	*USA*	*2015*	*19-24*	*DOI: 10.1557/opl.2015.725*	
3	*Membranes, electrodes, and membrane-electrodes assemblies analyzed before and after*	*R. Hiesgen*	*ECS Transactions*	*Volume 68, Issue 3*	*Electrochemical Society Inc*	*USA*	*2015*	*3-12*	*DOI: 10.1149/06803.0003ecst*	

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[5] A permanent identifier should be a persistent link to the published version full text if open access or abstract if article is pay per view) or to the final manuscript accepted for publication (link to article in repository).

[6] Open Access is defined as free of charge access for anyone via Internet. Please answer "yes" if the open access to the publication is already established and also if the embargo period for open access is not yet over but you intend to establish open access afterwards.

	operation by atomic force microscopy								
4	*Insight into the structure and nanoscale conductivity of fluorinated ionomer membranes*	*R. Hiesgen*	*Journal of the Electrochemical Society*	*Volume 161, Issue 12*	*Electrochemical Society Inc*	*USA*	*2014*	*F1214-F1223*	*DOI: 10.1149/2.0701412jes*	
5	*Influence of the distribution of platinum deposits on the properties and degradation of platinum-impregnated nafion membranes*	*R. Hiesgen*	*Journal of the Electrochemical Society*	*Volume 161, Issue 14*	*Electrochemical Society Inc*	*USA*	*2014*	*F1416-F1426*	*DOI: 10.1149/2.0531414jes*	
6	*Evaluation of reversible and irreversible degradation rates of polymer electrolyte membrane fuel cells tested in automotive conditions*	*P. Gazdzicki*	*Journal of Power Sources*	*Volume 327*	*Elsevier*	*The Netherlands*	*2016*	*86–95*	*http://dx.doi.org/10.1016/ j.jpowsour.2016.07.049*	*no*
7	*Design of Supported PtCo Electrocatalysts for Pemfcs*	*A.S. Arico*	*ECS Transactions*	*Volume 69*	*Electrochemical Society Inc*	*USA*	*2015*	*263-272*	*doi: 10.1149/06917.0263ecst*	
8	*Enhancement of Oxygen Reduction and Mitigation of Ionomer Dry-Out Using Insoluble Heteropoly Acids in Intermediate Temperature Polymer-Electrolyte Membrane Fuel Cells*	*A.S. Arico*	*Energies*	*Volume 8*	*Multidisciplinary Digital Publishing Institute*	*Switzerland*	*2015*	*7805-7817*	*doi:10.3390/en8087805*	*yes*
9	*Atomic Force Microscopy on Cross Sections of Fuel Cell Membranes, Electrodes, and Membrane Electrode Assemblies*	*R. Hiesgen*	*Electrochimica Acta*	*Volume 162*	*International Society of Electrochemistry*	*Switzerland*	*2015*	*86–99*	*http://dx.doi.org/10.1016/ j.electacta.2014.11.122*	
10	*Atomic force microscopy studies of conductive nanostructures in solid polymer electrolytes*	*R. Hiesgen*	*Electrochimica Acta*	*Volume 110*	*International Society of Electrochemistry*	*Switzerland*	*2013*	*292–305*	*http://dx.doi.org/10.1016/ j.electacta.2013.04.147*	
11	*Excavated Fe-N-C Sites for Enhanced Electrocatalytic Activity in the Oxygen Reduction Reaction*	*J. Lee*	*ChemSusChem*	*Volume 7*	*Wiley-VCH*	*Germany*	*2014*	*1289–1294*	*DOI: 10.1002/cssc.201301374*	
12	*Polydimethylsiloxane Treated Cathode Catalyst Layer to Prolong Hydrogen Fuel Cell lifetime*	*J. Lee*	*Catalysis Today*	*Volume 262*	*Elsevier*	*The Netherlands*	*2016*	*155-160*	*http://dx.doi.org/10.1016/j.cattod.2015.08.048*	

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13	Atomic layer deposition of ultrathin layered TiO2 on Pt/C cathode catalyst for extended durability in polymer electrolyte fuel cells	J. Lee	Journal of Energy Chemistry	Volume 25	Elsevier	The Netherlands	2016	258-264	http://dx.doi.org/10.1016/j.jechem.2016.01.010	
13	Surface Analytical Methods for the Development of Electrochemical Components of Polymer Electrolyte Fuel Cells	I. Biswas	ECS Transactions	Volume 58	Electrochemical Society Inc	USA	2013	1429-1444	doi: 10.1149/05801.1429ecst	
14	Atomic Force Microscopy Detection of Electronic Short-Circuits in Solid Polymer Electrolytes Fuel Cell Membranes after Accelerated Degradation	R. Hiesgen	ECS Transactions	Volume 58	Electrochemical Society Inc	USA	2013	1085-1096	doi: 10.1149/05801.1085ecst	
15	Liquid invasion from multiple inlet sources and optimal gas access in a two-layer thin porous medium	M. Prat	Transport in Porous Media	Volume 115	Springer	Luxembourg	2016	449-472	DOI: 10.1007/s11242-016-0630-1	
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A2: LIST OF DISSEMINATION ACTIVITIES

NO.	Type of activities[7]	Main leader	Title	Date/Period	Place	Type of audience[8]	Size of audience	Countries addressed
1	Workshop	Several partners	MATERIAL CHALLENGES FOR FUEL CELL & HYDROGEN TECHNOLOGIES	19 / 20 / 21 of September 2016	Grenoble, France	Fuel cell community		international
2	Workshop	All partners	2nd IMPACT Workshop on Degradation and Performance Issues of Fuel Cells for Automotive Applications	12th October 2016	Stuttgart, Germany	Fuel cell community	~40	international
3	Conference	DLR	World of Energy Solutions	10-12.10.2016	Stuttgart, Germany	Scientific Community, Industry, Politics		international
4	Conference	DLR	229th ECS MEETING	May 29 - June 2, 2016	San Diego, USA	Scientific Community		international
5	Conference	DLR	8. E-Motive Forum	7-8. Sept. 2016	Schweinfurt, Germany	Scientific Community, Industry,		national
6	Summer School	Several partners	IMPACT Summer School	September 22-23, 2016	Grenoble, France	Academia		european
7	Conference	DLR	Gordon Research Conference	August 7-12, 2016	Stonehill College Easton, MA, USA	Scientific Community		international
8	Conference	DLR	WORLD HYDROGEN ENERGY CONFERENCE	June 13th to 16th 2016	Zaragosa, Spain	Scientific Community		international
9	Workshop	Several partners	SecondAct Workshop	May 19th, 2016	Milan, Italy	Scientific Community		european
10	Conference	DLR, GIST	18th ISE Topical Meeting	March 8th to 11th, 2016	Gwangju, South Korea	Scientific Community		international

[7] A drop down list allows choosing the dissemination activity: publications, conferences, workshops, web, press releases, flyers, articles published in the popular press, videos, media briefings, presentations, exhibitions, thesis, interviews, films, TV clips, posters, Other.

[8] A drop down list allows choosing the type of public: Scientific Community (higher education, Research), Industry, Civil Society, Policy makers, Medias, Other ('multiple choices' is possible).

11	Conference	CNR	ENERCHEM-1	18-20 Feb. 2016	Florence, Italy	Scientific Community		national
12	Conference	CNR	Thermec 2016	29.05-03.06.2016	Graz, Austria	Scientific Community		international
13	Conference	CNR	ECS 228th Meeting	October 11-15, 2015	Phoenix, USA	Scientific Community		international
14	Workshop	Several partners	3rd International Workshop on Degradation Issues of Fuel Cells and Electrolysers	29/09/2015 to 01/10/2015	Santorini, Greece	Scientific Community, Industry		international
15	Conference	Several partners	EFCF 2015	30 June – 3 July 2015	Luzern, Switzerland	Scientific Community, Industry		international
16	Conference	CNR	ECS Conference	07/2015	Glasgow, UK	Scientific Community,		international
17	Conference	UAES, DLR	Material research Society (MRS) Spring Meeting	04/2015	San Francisco, USA	Scientific Community		international
18	Workshop	Several partners	IMPACT/IMPALA Public Workshop	02/2015	Toulouse, France	Scientific Community		international
19	Conference	Several partners	6th International Conference on "Fundamentals & Development of Fuel Cells"	02/2015	Toulouse, France	Scientific Community		international
20	Conference	Several partners	CARISMA	12/2014	Kapstadt, South Africa	Scientific Community		international
21	Symposium	GIST, UAES, DLR	3rd Ertl Symposium	11/2014	Berlin, Germany	Scientific Community		international
22	Workshop	UAES	4th International Workshop on Advanced Atomic Force Microscopy Techniques	03/2013	Karlsruhe, Germany	Scientific Community		international
23	Conference	DLR	Hydrogen and fuel cell conference	06/2013	Vancouver, Canada	Scientific Community, Industry		international
24	Conference	DLR	13th International Conference on Electrified Interfaces	06/2013	Prague, Czech Republic	Scientific Community		international
25	Conference	UAES, DLR	European Fuel Cell Forum	03/2013	Luzern, Switzerland	Scientific Community		international
26	Seminar lecture	UAES	Seminar lecture	2013	TTU Tallinn, Estonia	Academia		national
27	Conference	DLR	GDCh-Wissenschaftsforum	09/2013	Darmstadt, Germany	Academia		national
28	Conference	UAES	International Society of Electrochemistry	09/2013	Queretaro, Mexico	Scientific Community		international
29	Workshop	UAES	International Workshop PEM	10/2013	Trondheim, Norway	Scientific Community		international

30	Conference	DLR	European Conference of Applied Surface and Interface Analysis	10/2013	Fort Village, Italy	Scientific Community		international
31	Conference	UAES, DLR	224th ECS Meeting	10/2013	San Francisco, USA	Scientific Community		international
32	Conference	DLR	Spring Meeting of the German Physical Society	03/2014	Dresden, Germany	Academia		national
33	Workshop	DLR	International Workshop on PEMFC Stack and Stack Component Testing	06/2014	Stuttgart, Germany	Scientific Community		international
34	Conference	DLR, UAES	15th Topical Meeting of the International Society of Electrochemistry	04/2014	Niagara Falls, Canada	Scientific Community		international
35	Conference	UAES	Joint German-Canadian symposium on "Modeling and Diagnostics of Polymer Electrolyte Fuel Cells"	04/2014	Vancouver, Canada	Scientific Community		international
36	Conference	INPT, CEA	11th Euromech Fluid Mechanics Conference	09/2016	Sevilla, Spain	Scientific Community		international
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Section B (Confidential[9] or public: confidential information to be marked clearly)

Part B1

The applications for patents, trademarks, registered designs, etc. shall be listed according to the template B1 provided hereafter.

The list should, specify at least one unique identifier e.g. European Patent application reference. For patent applications, only if applicable, contributions to standards should be specified. This table is cumulative, which means that it should always show all applications from the beginning until after the end of the project.

TEMPLATE B1: LIST OF APPLICATIONS FOR PATENTS, TRADEMARKS, REGISTERED DESIGNS, ETC.					
Type of IP Rights[10]:	Confidential Click on YES/NO	Foreseen embargo date dd/mm/yyyy	Application reference(s) (e.g. EP123456)	Subject or title of application	Applicant (s) (as on the application)

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[9] Note to be confused with the "EU CONFIDENTIAL" classification for some security research projects.

[10] A drop down list allows choosing the type of IP rights: Patents, Trademarks, Registered designs, Utility models, Others.

Part B2

Please complete the table hereafter:

Type of Exploitable Foreground[11]	Description of exploitable foreground	Confidential Click on YES/NO	Foreseen embargo date dd/mm/yyyy	Exploitable product(s) or measure(s)	Sector(s) of application[12]	Timetable, commercial or any other use	Patents or other IPR exploitation (licences)	Owner & Other Beneficiary(s) involved
	Ex: New superconductive Nb-Ti alloy			*MRI equipment*	*1. Medical 2. Industrial inspection*	*2008 2010*	*A materials patent is planned for 2006*	*Beneficiary X (owner) Beneficiary Y, Beneficiary Z, Poss. licensing to equipment manuf. ABC*
New MEA	*New Low Loaded MEA designed for Automotive Applications. Incorporating corrosion resistant carbon and optimised stable ionomer*	*N*	*N/A*	*New Low Loaded MEA*	*Automotive*	*Immediate commercial availability after scale up*	*N/A*	*JMFC*

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[19] A drop down list allows choosing the type of foreground: General advancement of knowledge, Commercial exploitation of R&D results, Exploitation of R&D results via standards, exploitation of results through EU policies, exploitation of results through (social) innovation.

[12] A drop down list allows choosing the type sector (NACE nomenclature) : http://ec.europa.eu/competition/mergers/cases/index/nace_all.html