FINAL PUBLISHABLE SUMMARY REPORT

FCH JU GRANT AGREEMENT NUMBER: N° 300081

PROJECT ACRONYM: ELECTROHYPEM

PROJECT TITLE: Enhanced performance and cost-effective materials for long-term operation of PEM water electrolysers coupled to renewable power sources

FUNDING SCHEME: Collaborative Project - FCH JU GRANT AGREEMENT - SP1-JTI-FCH.2011.2.7 Innovative Materials and Components for PEM electrolysers

DATE OF LATEST VERSION OF ANNEX I AGAINST WHICH THE ASSESSMENT WILL BE MADE: 05 April 2012

PERIODIC REPORT: **Final Publishable Summary Report**

PERIOD COVERED: *36 months* **from** 01-07-2012 **to** 30-06-2015

NAME, TITLE AND ORGANISATION OF THE SCIENTIFIC REPRESENTATIVE OF THE PROJECT'S COORDINATOR:

Dr Antonino Salvatore Aricò, CNR-ITAE, CONSIGLIO NAZIONALE DELLE RICERCHE - Italy

TEL: +39 090 624237

FAX: +39 090 624247

E-MAIL: arico@itae.cnr.it

PROJECT WEBSITE ADDRESS: http://www.electrohypem.eu/

TABLE OF CONTENTS

A. FINAL PUBLISHABLE SUMMARY REPORT

EXECUTIVE SUMMARY

The overall objective of the ELECTROHYPEM project (http://www.electrohypem.eu) was to develop cost-effective components for polymer electrolyte membrane (PEM) water electrolysers with enhanced activity and stability in order to reduce stack and system costs and to improve efficiency, performance and durability. In accordance with the specific area on Hydrogen Production & Distribution and the call topic SP1-JTI-FCH.2011.2.7 Innovative Materials and Components for PEM electrolysers, the project has dealt specifically with cost-effective and enhanced durability components for PEM electrolysers amenable to be integrated with renewable power sources. The activities in the project were oriented towards both short and long term innovation.

Polymer electrolytes developed in the project included novel reinforced short-side chain chemically stabilised perfluorosulphonate ionomers and sulphonated hydrocarbon membranes, as well as composite or reinforced membranes. The objectives for these membranes were high ion conductivity, wide operating temperature range (up to 150 °C), better resistance than conventional Nafion membranes to H_2-O_2 cross-over and reduced mechanical degradation under high pressure operation. Low noble-metal loading nanosized mixed-oxides oxygen evolution (OER) and supported hydrogen evolution (HER) electrocatalysts were developed together with novel non-precious oxygen evolution electrocatalysts. Large area membrane-electrode assemblies (MEAs) based on these components were validated in PEM electrolyser prototypes. The stack was integrated in a system and assessed in terms of durability under steady-state operating conditions as well as in the presence of current profiles simulating intermittent conditions.

The main project milestones were the achievement of membrane conductivity better than 200 mS/cm & H_2 permeation rate <6.25 10^{-5} cm s^{-1} at ambient pressure or <1% H_2 concentration in the oxygen stream at high differential pressure , low overpotentials for anode/cathode reactions (OER overpotential < 0.1 V vs. E thermoneutral, HER overpotential < 0.05 V vs. RHE at 1 A cm^{-2}), noble metal loading <0.5 mg cm^{-2}; large area (130 cm^2) MEAs characterized by current densities better than 1 A cm^{-2} @ 1.6 V/cell and 3 A cm^{-2} @ 1.8 V/cell, degradation <10 μV/h and stacks with low energy consumption ~3.5 kWh/Nm^3 H_2 at a hydrogen production rate > 1.2 Nm^3 h^{-1} with stack efficiency better than 80 % LHV. The achieved results demonstrate the progress beyond the state of the art. A significant aspect of the project was also dealing with the implementation of a system around the stack for assessing the dynamic behavior, in particular testing the stack under simulated renewable energy sources (RES) current profiles.

Dissemination activities included:

- Research publications in peer-reviewed journals
- Meetings, conferences
- Organisation of dedicated workshops
- Project Web-site.

In total 7 journal articles have been published in peer-reviewed scientific journals and 15 presentations (oral or posters) delivered at international conferences. An Electrohypem workshop on PEM electrolysis was organized with project results presentation.

A list of planned dissemination activities to be continued after the end of the project has been provided as well as exploitable foregrounds & potential impact.

SUMMARY DESCRIPTION OF PROJECT CONTEXT AND OBJECTIVES

Hydrogen is a promising energy carrier that can be produced by various technologies. Electrolysis of water using renewable energy sources (RES) is an essential contributor, due to the relatively high efficiency and the production of high purity hydrogen. Overcapacities in electricity production from renewable power sources can be suitably addressed by converting the surplus of electrical energy into hydrogen. Renewable energy sources and electrolysers can thus be matched directly or through the grid with a high coupling efficiency. PEM electrolyser can provide superior grid-balancing service. However, there are several challenging issues, such as substantial energy losses, materials degradation and cost, that must be conveniently addressed to achieve large-scale deployment of this technology.

The most relevant issue for the present PEM electrolysis devices is related to the use of expensive perfluorinated membranes, noble metal catalysts, and titanium-based current collectors. In order to promote a wide scale decentralised hydrogen production using PEM electrolysers for direct coupling with renewable power sources and to provide grid-balancing service, cost-effective and compact devices must be developed with high durability (approaching 100 khrs) and capability to producing hydrogen at very high efficiency (>80%) and reasonable pressure (>30 bar).

For the electrolysis process, the oxygen evolution electrode is the main source of overpotential. But, at high current density which is required to decrease capital costs, also the contribution to energy losses of the polymer membrane electrolyte resistance is significant. Moreover, the Titanium bipolar plates affect significantly the cost of PEM electrolysis stacks. In addition, mechanical, chemical and electrochemical stability play an important role. It is therefore important to develop novel and optimized materials to minimize energy loss and enhance system durability. Before this technology can reach a full scale production, also problems related to the high cost must be solved.

The focus of the project was thus on low-cost electrocatalysts, low-noble metal loading electrodes, cheap bipolar plates and robust membrane development. The project addressed the development of PEM electrolysers based on such innovative components for residential applications in the perspective of a suitable integration with renewable power sources. The aim is to contribute to the road-map addressing the achievement of a wide scale decentralised hydrogen production infrastructure.

The objectives of Electrohypem were to carry out the management, scientific, technical and dissemination activities reported in the Description of Work in an integrated manner and to achieve the milestones and deliverables planned according to the project schedule. The specific objectives of the various WPs are reported below:

WP1

- To coordinate negotiation of the consortium agreement, its finalisation and signature;
- To distribute project funds in a timely manner;
- To coordinate the scientific and technical activities of the project and interactions between the partners and between work packages;
- To provide communication tools adapted to the needs of the partners and promote their use.
- To coordinate preparation and finalisation of deliverable reports, technical project progress reports and financial reports.
- To interface effectively between the European Commission officers, the project and its partners.
- To establish a mutual understanding and confidence between project partners;
- Organising progress meetings (two per year); chairing technical, management meetings.
- Ensuring timely delivery of samples/devices, deliverables, reports and cost statements (including budgetary overviews).

WP2

- To define characterisation and test protocols for the assessment of performance, efficiency, durability and cost-effective characteristics of the newly developed polymer electrolyte membrane water electrolyser (PEM WE) components
- Describe a set of protocols for the PEM electrolyser in relation to duty cycle experiments and testing in relation to interfacing with specific intermittent renewable power sources.

- Establish a benchmark of baseline components against which progress can be assessed in terms of durability, performance and cost.

- Identify appropriate accelerated stress test (AST) protocols

WP3

- Developing innovative PEM electrolyser membranes. Cost-effective short side chain perfluorosulphonic acid (PFSA) and hydrocarbon or heterocyclic membranes in order to achieve:
    - high proton conductivity (higher than $2 \cdot 10^{-1}$ S cm^{-1}) at the specific targeted temperatures and membrane related ohmic loss of 0.05 V at 2 A cm^{-2} for a reinforced membrane with thickness > 50 μm .

  - low gas cross-over for both H_2 and O_2 i.e. H_2 permeation rate <6.25 10^{-5} cm s^{-1} or lower than 1 mA cm^{-2} equivalent current density at the targeted temperatures and ambient pressure or below 1-2 vol.% at relevant pressures and high current densities e.g. 1 A cm^{-2}.
- operating temperature range for unfilled membranes from R.T. to 90°C and up to 150 °C under pressure for composite membranes.

WP4

- Enhancing the stability, reduce the cost and increase the performance of PEMWE electrocatalysts.
- Reduce anode overpotential vs. thermoneutral potential for OER to less than 100 mV (IR-free) at 1 A cm^{-2} and cathode overpotential vs. RHE to less than 50 mV (IR-free) at the same current.
- Develop a Pt-based recombination catalyst able to reduce H_2 content in O_2 stream to less than 1%.

WP5

- Validation in terms of stability and performance of the novel membranes in combination with enhanced performance and durability catalysts in practical MEAs under operation in single cell at the targeted temperatures and pressures.
- Reduction of noble metal loading to less than 0.5 mg cm^{-2}.
- Reduction of MEA performance degradation to less than 15 μV/h.
- Achieve performance in terms of current density better than 1 A cm^{-2} @ 1.6 V/cell and 2 A cm^{-2} @ 1.8 V/cell.
- Scaling-up of MEAs for provision to stack assembling. Practical MEA geometrical area ≥ 100 cm^2.

WP6

- Assessment of the new developed components in verification units i.e. PEM electrolyser stacks for testing under realistic operating conditions and validation with respect to the project objectives.
- Full stack PEM electrolyser of rated hydrogen production capacity of 1 $Nm^3 h^{-1}$ operating at an efficiency equal or better than 80% vs. the Lower Heating Value (LHV) of hydrogen in a wide temperature range with energy consumption lower than 4 kWh/Nm^3 H_2 at the nominal production capacity.
- Assessment of components durability in a durability test under practical operating conditions at with a terminal voltage increase per cell < 15 μV/h.
- Implementation of a system (BoP) around the stack for testing under simulated renewable energy sources (RES) current profiles.

WP7

- Provide the maximal dissemination of the results and implementation of developed PEM electrolyser components, stack and system in industry by using different information and dissemination tools such as, workshops, project web site, etc.
- Contribute to public understanding of scientific research and technological development in Europe by means of articles and conferences.
- Extension of public dissemination through dedicated web-site.
- Evaluation of industrial scalability of the production process for down-selected components and assessment of additional steps required before product commercialisation.

A.3 DESCRIPTION OF THE MAIN S&T RESULTS/FOREGROUNDS

I. WP1. PROJECT MANAGEMENT – WPL: CNR-ITAE

The activity of WP1-Project management was essentially addressed to provide efficient project coordination and management to support the partners in their achievement of the project objectives, to interface with the FCH JU and to provide tools for communication between partners inside the consortium. This was achieved by the development of web-based tools (Intranet), as well as representing the project to outside audiences through the public web site (http://www.Electrohypem.eu).

The coordination activity has been specifically addressed to ensure that each of the technical work packages started effectively and in a timely manner, so as to avoid any delays with respect to the project schedule.

Significant efforts were addressed to allow completion and submission of all deliverables due by the end of the project as well as to the achievements of the general objectives of the project and the specific milestones.

The project rapidly acquired a public face through the implementation of the project website, as well as a user-friendly and highly efficient shared workspace where project documents were uploaded and stored. These sites are maintained and updated on a continuous basis.

Steering committee and technical progress meetings have been attended by the partners (a list of these meetings and the relative agendas are reported in the progress reports), with active participation and discussion especially in relation to protocols, methods, activities, results, achievements and dissemination.

Consortium agreement was completed and signed in line with the commencement of the project.

Technical and Financial reporting have been completed.

An amendment to the Grant Agreement was needed in the second period to take into account the following aspects:

1. An UTRO from Universite Montpellier 2 to Universite Montpelier
2. ITM Research was introduced as a third party making available resources for ITM Power trading

Moreover some legal representatives for some beneficiaries, changed during the second period and these changes were included in the amendment letter.

No other deviations or changes have been occurred in the Consortium. The project was executed in-line with the project planning in the DoW.

The coordinator has represented the project at international conferences, in particular at the FCH-JU review days in Brussels in 2012, 2013, 2014.

II. WP2. SPECIFICATION, PROTOCOLS & COST ANALYSIS – WPL: CNR-ITAE

In the framework of the Electrohypem project, specifications and protocols (WP2) for assessing polymer electrolyte membrane water electrolyser components and devices such as membranes, catalysts, MEAs and stacks have been delivered. These are available to the public through the project web-site (http://www.electrohypem.eu). Characterisation and testing protocols for ex situ and in situ characterisation of innovative membranes, catalysts and supports, MEAs and stacks have been defined in Workpackage 2. In particular, these protocols allow for a comparison of characterisation data between the partners, and enable a homogeneous screening and evaluation of the newly developed components. Baseline components have been identified against which progress can be assessed.

During the second period of the Electrohypem project, some specifications and protocols for assessing polymer electrolyte membrane water electrolyser components and devices such as membranes, catalysts, MEAs and stacks have been further refined according to the feed-back provided by the other RTD WPs and the observations raised during the mid-term meeting. Two deliverables D2.1 and D2.2 were improved and re-submitted. These are available to the public through the project web-site (http://www.electrohypem.eu). Beside conventional testing protocols, specific protocols were addressing the dynamic behaviour of the PEM electrolyser such as load and on-off cycles, duty cycle experiments using current profiles simulating the behaviour of specific intermittent renewable power sources.

An analysis of stack capital costs and of the perspectives of cost reduction in relation to the development of new stack components and system simplification was carried out (D6.2). This activity was shared between WP2 and WP6 and included an analysis of the projected cost for hydrogen produced by PEM electrolysis and recyclability of the stack components at the end of life.

A summary of cost analysis results is provided below:

Regarding catalysts and membrane, in a standard stack configuration, these are responsible respectively for a 5% and a 8% of the total stack cost. Being capital costs inversely related to the operating current density, a reduction between 6 and 12% of the total costs for a standard PEMWE stack is obtained using the new catalysts and membranes developed within the project and the Electrohypem operating conditions (current density of 3 A cm^{-2}). This corresponds to a decrease of 4-7% of the cost of the entire system (based on a standard stack architecture) that can be achieved by introducing new ElectroHyPEM membranes and catalysts.

ElectroHyPEM specific costs breakdown is shown in figures 1 and 2 both at stack and system level for standard configuration but operating at 3 A/cm^2 with regular catalyst loading. These figures do not yet include the bipolar plates developments that are discussed successively.

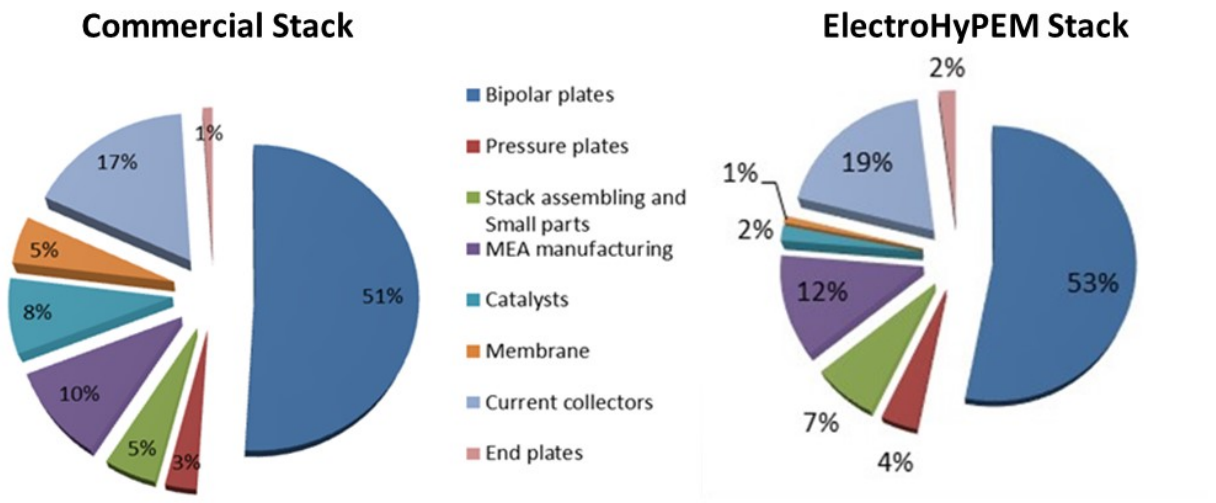


Fig. 1 Capital stack costs for a standard commercial PEM electrolyser stack vs a PEM electrolyser stack with similar design but containing Electrohypem MEAs (TRE).

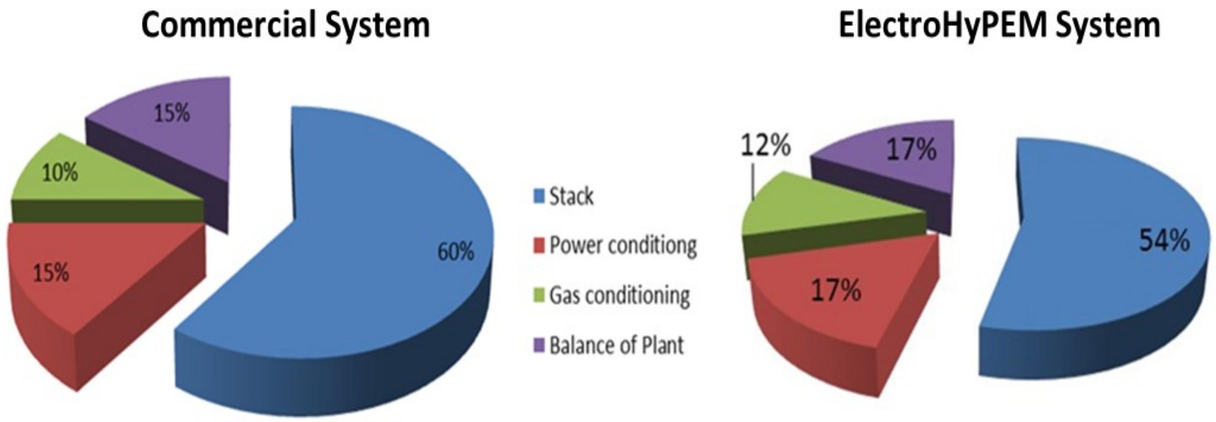


Fig. 2 Capital stack costs for a standard commercial PEM electrolyser system vs a PEM electrolyser system based on a similar stack architecture but containing Electrohypem MEAs (TRE).

The impact of changing stack architecture and type of bipolar plates on the electrolyser cost was also considered. In Electrohypem, the inside of the electrolyser stack has been reconfigured to eliminate expensive machining costs. The traditional "bipolar plates" have been replaced for lower costs components and injection moulded parts. A cost breakdown of the newly reconfigured stack is given in figure 3.

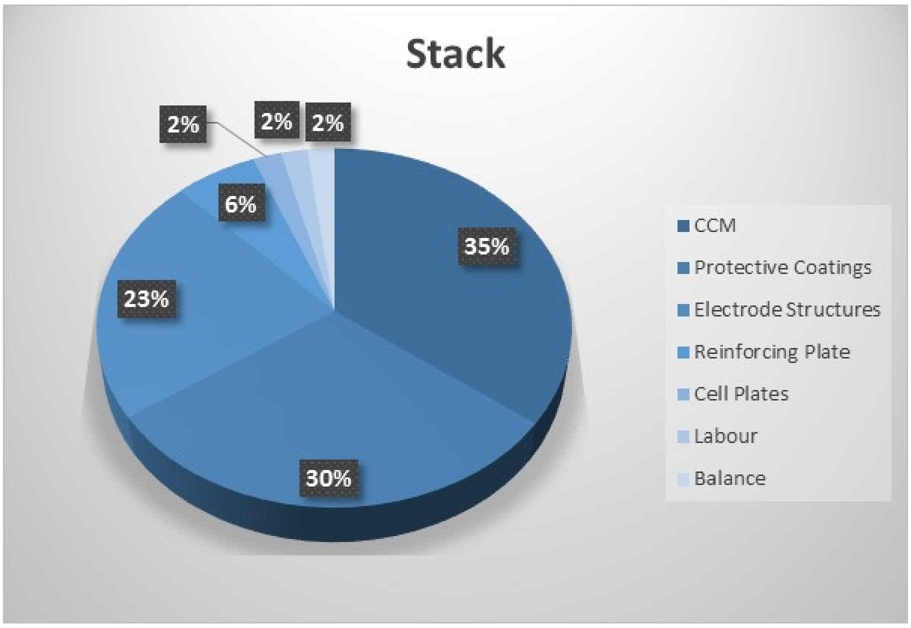


Fig. 3 Costs breakdown of the newly designed stack (ITM)

As the cost of the plates is now significantly lowered, the catalyst-coated membrane (CCM) becomes a more dominant cost and the coating of electrode structures becomes the second most expensive part of the stack "behind the CCM". The coating is necessary to extend the lifetime of the stack to the requisite 100 khrs of operation. A low cost coating has been developed based on sol-gel chemistry that significantly lowers the cost of coating the internal cell components (bipolar plates). The development of the sol-gel coating (still at R&D stage) has the potential to lower the cost of coating the components by 48%. This will lower the stack costs by 14%. The combination of CCM savings and coatings developed in ElectroHyPEM will finally lower the cost of a production stack by 26%. By increasing the current density it is possible to increase these savings considerably. In Electrohypem, for some specific MEA configuration, it was demonstrated a very low degradation rate that would indicate a perspective of 100 khrs life-time which favourably compares to the life-time of standard systems that at the present is not larger than 40 khrs. Increasing the lifetime from 40 khrs to 100 khrs will significantly lower the maintenance costs of the electrolyser. Traditionally in a twenty year lifetime hydrogen refuelling station, the stack(s) will need to be replaced three times. Increasing the lifetime to 100 khrs will lower this to one replacement (assuming 80 % utilisation in both cases). This will significantly reduce the maintenance cost of stack replacement by 50%. In addition, the cost of hydrogen from an electrolyser is not only based on electricity and water costs but the capex cost amortised over the lifetime of the product. As we lower the amortisation rate, by increasing the lifetime, the cost of hydrogen will come down.

III. WP3 INNOVATIVE MEMBRANES - WPL: SOLVAY

In the first phase of the project, a significant number of polymer electrolyte membranes varying in terms of composition, equivalent weight, thickness, filler and reinforcement have been developed at Solvay, ITM, CNRS and CNR-ITAE. All materials have been screened in terms of proton conductivity, cross-over, mechanical and hydrolytic stability. The activity of the first period in WP3 essentially addressed the screening of the most promising membranes, on the basis of the relevant properties individuated in WP2, and were validated in MEAs and short stacks in the second phase of the project. Extruded and reinforced membranes made of short side chain perfluorinated ionomer with a thickness of about 100 µm have been developed by Solvay for PEMWE applications. Proton conductivity better than 200 mS cm^{-1} at 80 °C has been obtained. In parallel, hydrocarbon-based, cross-linked polymers have been developed by ITM in the framework of the Electrohypem project. A number of new monolithic and composite membranes have been produced and characterised. Promising results have been obtained for some monolithic membranes with regard to meeting the proton conductivity target of 200 mS cm^{-1} at ≥ 80 °C. CNRS has focused on reinforcing Aquivion membranes using electrospun fibers of polysulfone and polybenzimidazole for high temperature applications covering the temperature range up to 140 °C under pressure, and on the investigation of sulfonated polybenzimidazole membranes as a low cost alternative to PFSA. Hydrogen cross-over for these polymer electrolytes was lower than 1 mA cm^{-2} equivalent current density. The obtained results favourably compare to the quantitative milestones (MS1 conductivity >0.2 S cm^{-1}, MS2 H_2 cross over < 1 mA cm^{-2}, operation up to 150 °C) defined in the project for the membrane characteristics.

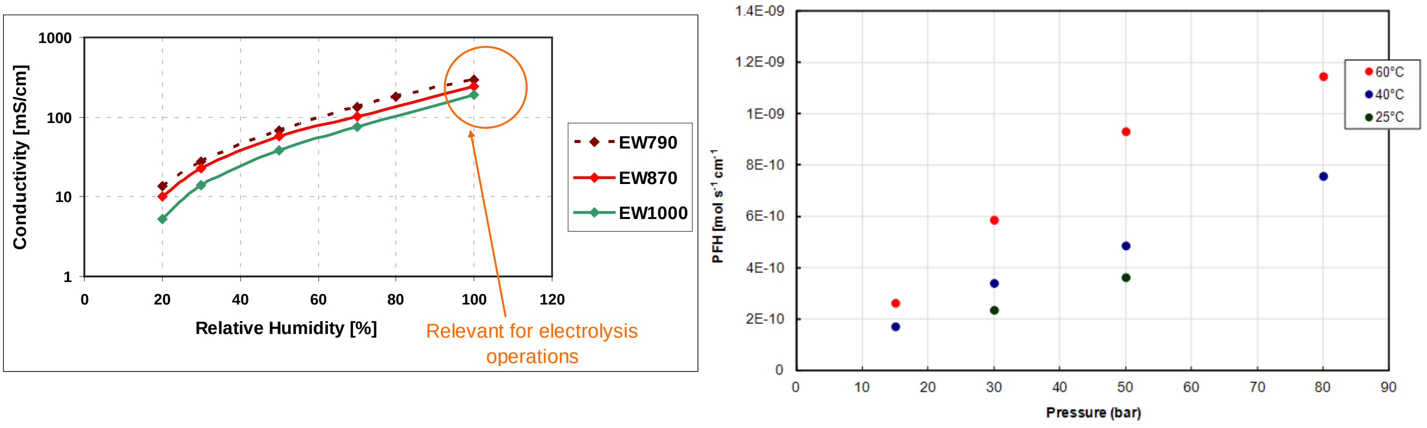


Fig. 4 Conductivity vs. R.H. for Solvay Aquivion polymers and permeation factor (PF_H) of hydrogen for the Aquivion membrane as a function of pressure (SOLVAY, ITM).

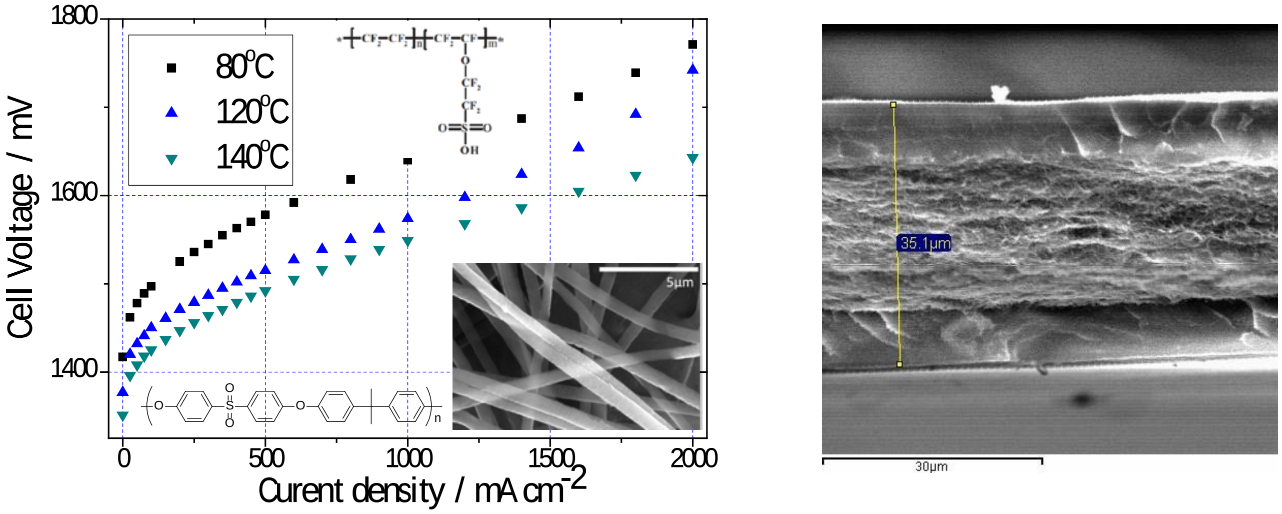
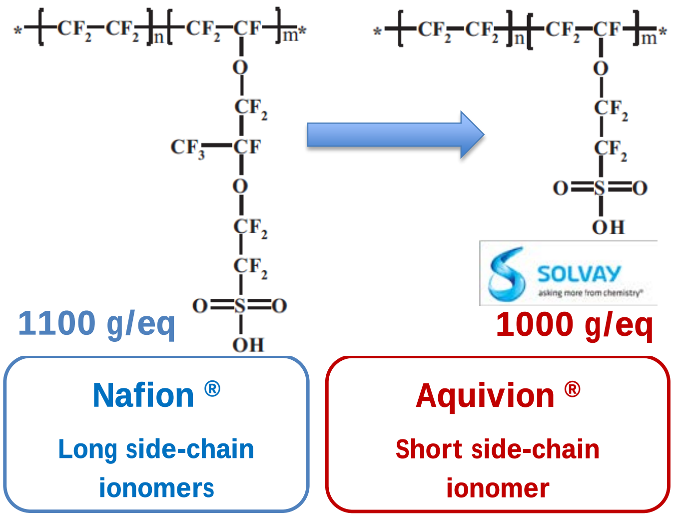


Fig. 5 High temperature electrolysis operation of a composite membrane of Aquivion, electrospun polysulfone nanofibre mats and cross-section of a reinforced Aquivion membrane (CNRS, SOLVAY).

In the second phase of the project, most of the efforts on polymer electrolyte membranes were focused on the down-selected short-side chain Solvay Aquivion perfluorosulphonic membrane. Optimisation was carried out at Solvay and CNRS in terms of equivalent weight, thickness, filler and reinforcement.



The Solvay Aquivion® membrane has higher conductivity, larger crystallinity and higher glass transition temperature than Nafion® as well as lower gas-cross over in the presence of reduced thickness.

Fig. 6 Main properties of the down-selected Aquivion membrane (SOLVAY)

A scaling-up of the selected Aquivion membrane was carried out. After assessing the scaled-up membrane in terms of proton conductivity, cross-over, mechanical and hydrolytic stability, these were validated in large area (130 cm^2 active area) MEAs and short stacks in the second phase of the project. Following the positive results, the Aquivion membranes were produced in suitable amounts for MEAs manufacturing and assembling the full stack. The selected polymer electrolytes were extruded and reinforced membranes made of short side chain perfluorinated ionomer with a thickness of about 100 µm developed at Solvay. Proton conductivity better than 200 mS cm^{-1} at 80 °C has been obtained. Gas cross-over, determined in a real system under differential pressure 15 and 30 bars and before the recombination catalyst, was lower than 1% H_2 in the oxygen stream. CNRS has continued in the second period their activity on reinforcing Aquivion membranes using electrospun fibers of polysulfone and polybenzimidazole for high temperature applications covering the temperature range up to 140 °C under pressure. The specific project milestones were also achieved for the scaled-up membranes (MS1 conductivity >0.2 S cm^{-1}, MS2 H_2 cross over < 1 mA cm^{-2}, operation up to 150 °C under pressure).

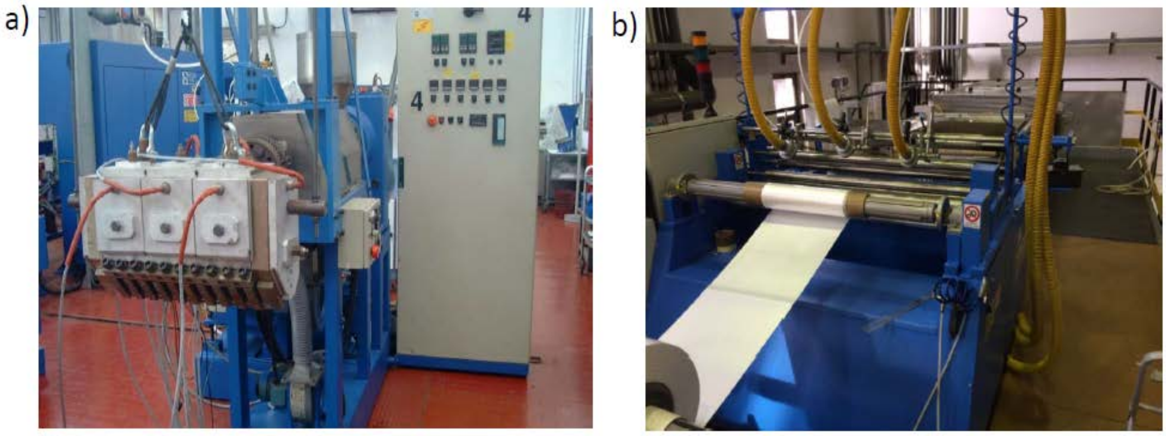


Figure 7 a) extrusion head and b) hydrolysis line of Aquivion E100-09S PFSA (SOLVAY)

Regarding, the membrane interface with electrodes, a slightly lower series resistance was obtained for the electrolyser equipped with Aquivion® compared to Nafion® . This is clearly related to the higher conductivity of the short-side chain perfluorosulphonic membrane since the thickness of the two membranes is comparable; it is essentially related to the smaller equivalent weight (1000 vs. 1100 g/eq). Whereas, a lower polarization resistance recorded for the Aquivion® interface compared to Nafion® is certainly arising from different electro-kinetic features associated with the catalyst ionomer electrolyte interface.

IV. WP4 ENHANCED ELECTRO-CATALYSTS – WPL: CNR-ITAE

New catalytic formulations as well as modified conventional electro-catalysts were developed in WP4 to find appropriate synergisms for enhancing the catalytic activity, and to address cost-effectiveness by reducing noble metal loading or developing non noble metal formulations. The activity of the first period in WP4 was mainly addressing the down-selection of the most performing catalysts. The research activities were regarding both supported and unsupported electrocatalysts.

Bare and doped Ti-oxide supports have been prepared at CNR-ITAE by using various synthesis routes. A crystalline Anatase phase was obtained for most of the doped and undoped Ti-oxide supports after thermal treatment in a range 300° - 600 °C. Rutile structure was observed for treatments at higher temperatures. Magneli phase was obtained after reduction at high temperature in hydrogen (1050 °C). BET surface area decreased passing from bare TiO_2 to Ta-doped TiO_2 and further decreased in the presence of Nb doping. Crystalline materials with crystallite domain sizes of about 3-4 nm were obtained upon thermal treatment at 400 °C for the Anatase phase. The corresponding BET surface area for bare TiO_2 was around 250 m^2/g whereas it decreased to 175 m^2/g for the doped support. Ti-suboxides with Magneli phase, obtained by high temperature reduction, were characterised by low surface area. For oxide supported catalysts, it was necessary a suitable coverage of the support with the catalytic conductive phase (e.g. IrO_2) to enhance electronic percolation on the surface.

Unsupported IrO_2 catalyst developed at CNRS shows excellent activity throughout the temperature range, and its simple method of preparation is highly promising with regard to scaling-up. Solid solution of nanosized Ir and Ru oxides have been prepared at ITM resulting in excellent performance at low temperatures (55°C) and suitable stability. The most promising CNR-ITAE formulations were IrRuOx anode electrocatalyst and Pt/C cathode catalyst. (Ir,Ru)Ox electrocatalyst based on the crystallographic structure of IrO_2 but with ruthenium present in solid solution allows to harness the extra electrochemical activity of ruthenium oxide while avoiding its dissolution which occurs if used alone or in mixtures via oxidation to RuO_4. Good stability was obtained with $Ir_{0.7}Ru_{0.3}O_x$ prepared by a molten salt procedure (Adams process), Pt/C by a sulphite complex method process (CNR-ITAE) and IrRuOx by a hydrolysis process (CNRS). All appeared suitable for scaling up. The amount of catalyst obtained in a single batch preparation was significantly larger than that required for the full stack.

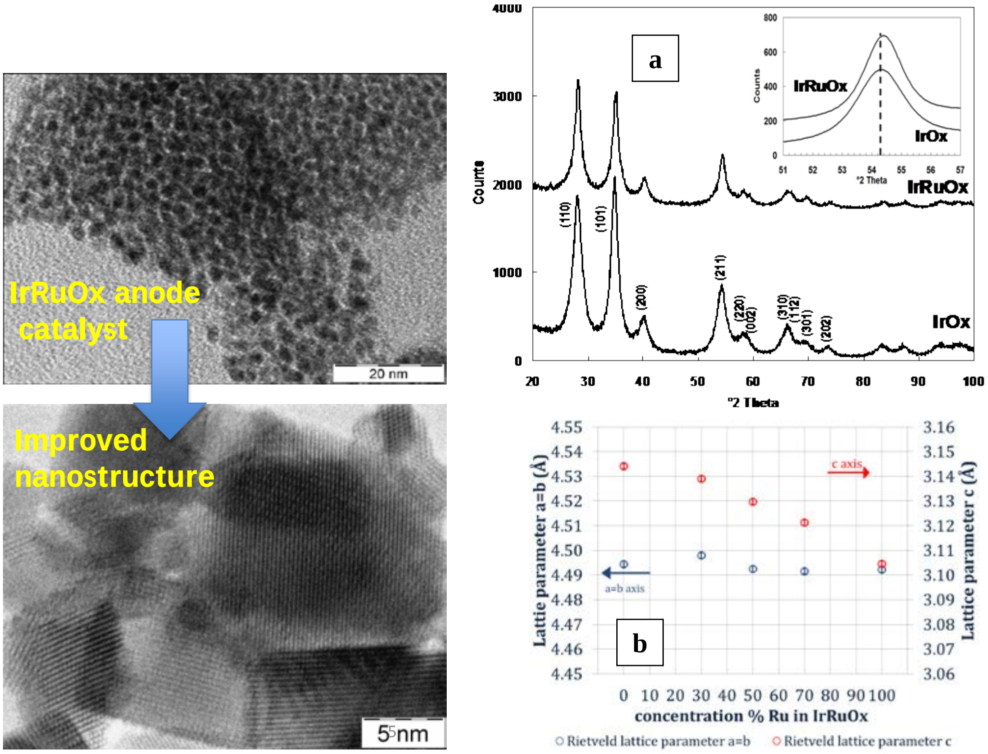


Fig. 8 From amorphous to crystalline nanostructure of Ir-oxide CNR-ITAE catalysts a) XRD patterns of the CNR-ITAE IrOx and $Ir_{0.7}Ru_{0.3}O_x$; (b) crystallographic parameters for IrRuOx vs. Ru content (ITM).

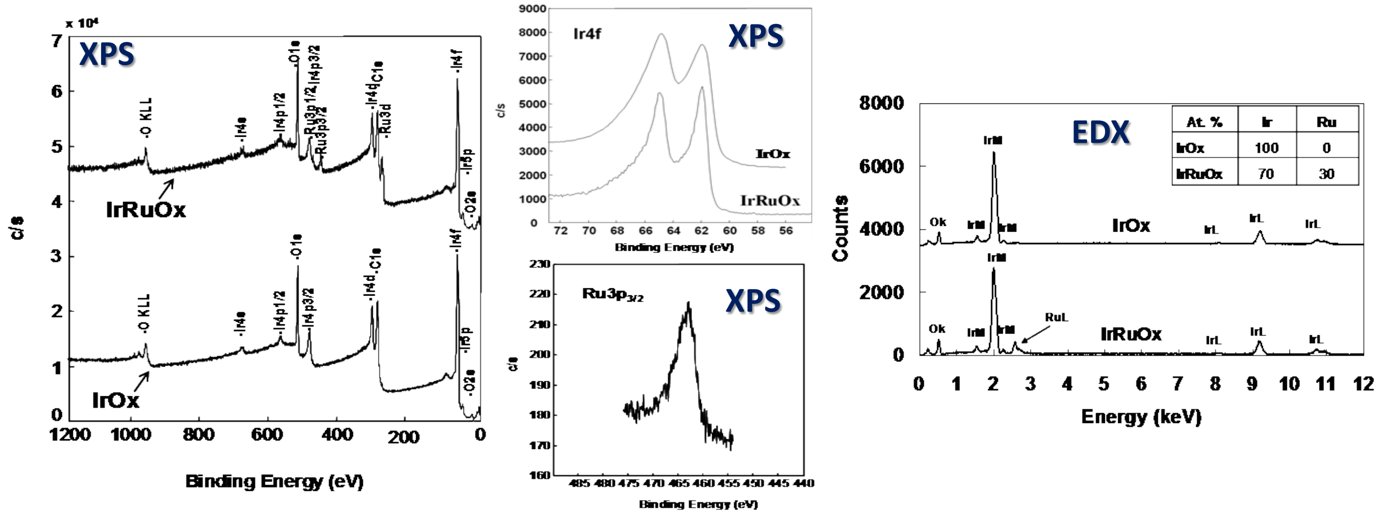


Fig. 9 Surface (XPS) and bulk (EDX) chemistry of CNR-ITAE IrOx and $Ir_{0.7}Ru_{0.3}O_x$ catalysts

Nanosized Pt (supported on carbon black) and Ir-oxide based electrocatalysts with enhanced mass activity have been developed for hydrogen and oxygen evolution reactions (HER and OER), respectively, in PEM electrolysers. Overpotentials for the OER vs. the thermoneutral potential (1.48 V) at 1 and 2 A cm^{-2} of 60 and 83 mV have been recorded at CNR-ITAE. The value achieved at 1 A cm^{-2} represents a 40% improvement over the project milestone (MS4) regarding anode performance. Whereas for the HER, the recorded overpotential at 1 A cm^{-2} was about 50 mV vs. the reversible hydrogen potential that is comparable to the quantitative project target for the cathode (MS5).

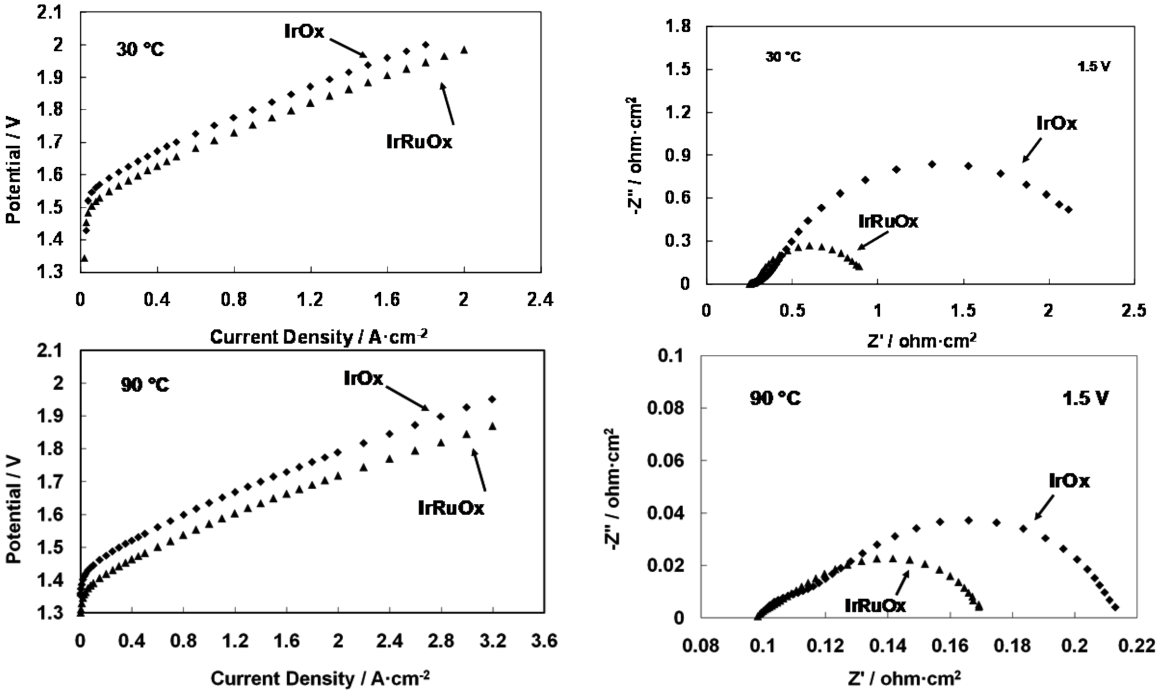


Fig. 10 Comparison of polarization curves and Nyquist plots of IrOx- and $Ir_{0.7}Ru_{0.3}O_x$-based electrolyser cells at 30 °C and 90 °C (CNR-ITAE).

Electrochemical experiments of the Pd/C and Pt/C HER catalysts showed modest performance for Pd compared to Pt at the same level of noble metal loading. This may be due to the occurrence of strong bonds between adsorbed hydrogen species on the surface and Pd catalytic sites. Such effect reduces the desorption rate of H_2 molecules.

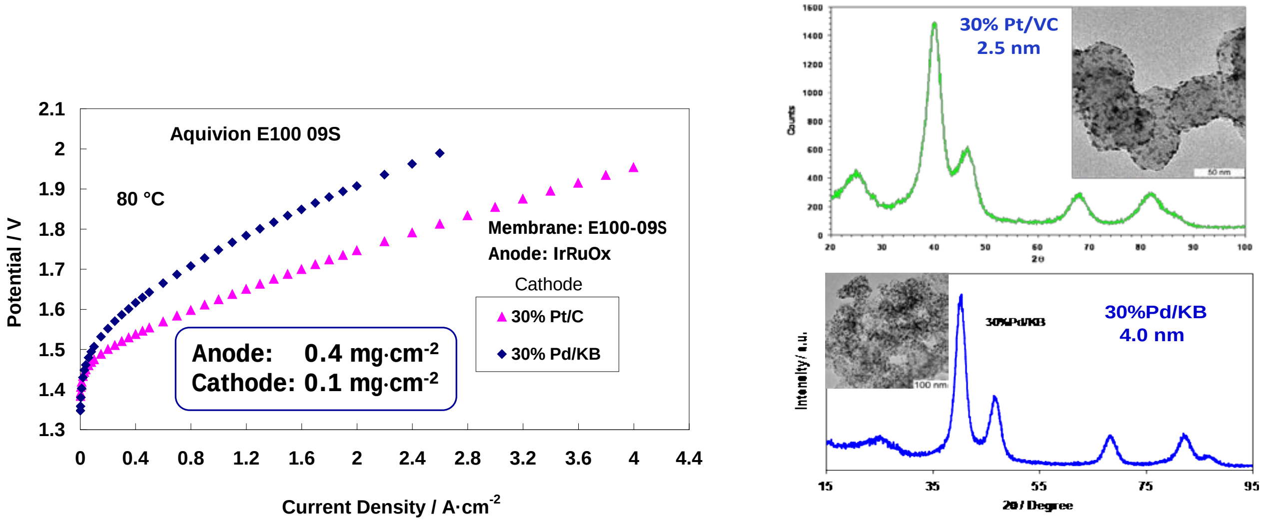


Fig. 11 Polarisation curves of a PEM electrolyser equipped with ITM IrRuOx anode and Pd/C and Pt/C HER catalysts developed at CNR-ITAE.

These catalysts combined to the Aquivion ionomer (SOLVAY) have provided excellent performance, in line or better than the project targets (MS4, MS5). An assessment of the noble metal catalysts prepared in the project was carried out by SOLVAY. The performances achieved with the Electrohypem catalysts favourably compared to benchmark commercial catalysts. Good perspectives were also envisaged to significantly decrease the noble metal loading (see WP5).

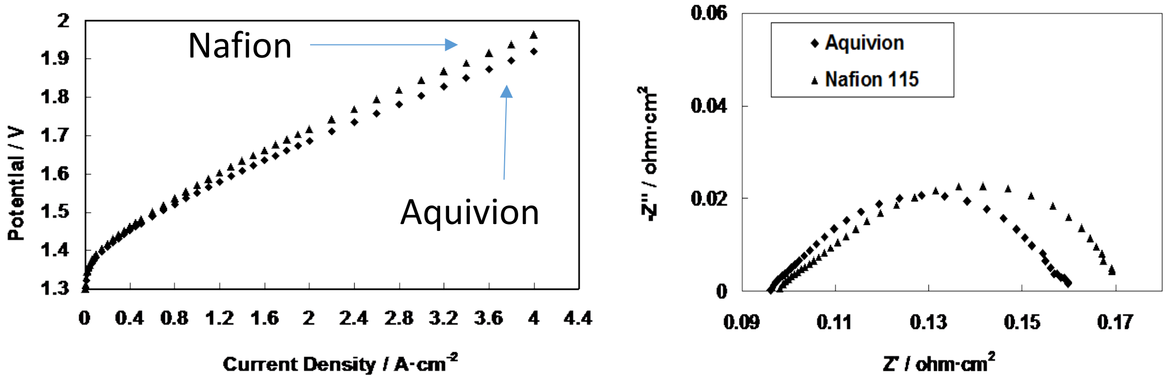


Fig. 12 Comparison of the water electrolysis polarization behavior and ac-impedance spectra (1.5 V) for CNR-ITAE electrocatalysts in Aquivion® and Nafion®-based MEAs at 90°C.

The activity on innovative catalytic formulations continued in WP4 in the second phase focusing on the further enhancement of catalytic activity and stability of Ir-Ru oxide solid solutions for the anode and carbon supported Pt and Pd for the cathode formulations down-selected from the first phase. Further efforts were addressed to achieve cost-effectiveness for these catalysts formulations by reducing noble metal loadings. Scaling-up processes were thus developed for the down-selected catalytic formulations in the range of tens of grams. The properties of the up-scaled formulations were thus assessed against the small-scale catalysts batches, and further tests were carried out under MEA configuration for a quality control. These were thus supplied for assembling and testing in short stacks and full stacks.

An explorative study on non-noble metal oxygen evolution catalysts was carried out (CNRS). A family of materials were prepared with molar ratios, M/Co=2 and M/Co=4, with the general formula of $MxNyCo_2O_4$, where M and N, are Ni, Co, Mn or Cu. These were prepared using co-axial electrospinning and evaluated for oxygen evolution. Non noble metal formulations activity continued in the second period by assessing these formulations at cell level. The most promising results are shown below.

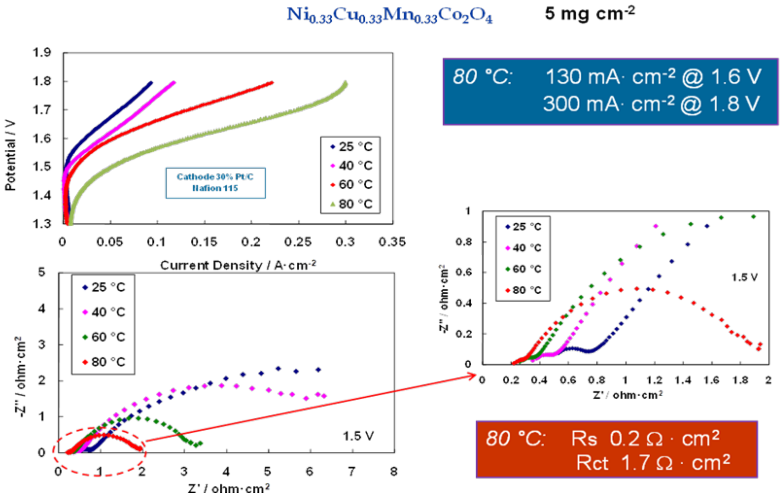


Fig. 13 Performance of non noble metal anodes developed at CNRS and tested in MEAs at CNR-ITAE.

To summarize the electrocatalysts part, main scientific and technical achievements in WP4 were concerning with the development of designed electro-catalysts capable of achieving the targeted electro-catalytic activity under specific operating conditions according to the project milestones. The target electrocatalytic activity (MS4-MS5, reduce overpotential vs. for OER and HER to less than 50-100 mV at 1 A cm^{-2}) were also achieved for the catalyst produced in large batches. Other relevant aspects were the reduction of the noble metal loading versus conventional formulations, the development the individuation of anodic formulations based on non-noble metal catalyst with promising performance characteristics. Performance of non-metal formulations was relatively modest compared to IrOx and IrRuOx catalysts. This was essentially due to an increase of series resistance related to electronic conductivity and polarization resistance (catalytic effect). A limiting current density of 300 mA cm^{-2} was recorded at terminal cell voltage of less than 1.8 V for the $Ni_{0.33}Cu_{0.33}Mn_{0.33}Co_2O_4$ spinel structure. In terms of system capital costs, since the non noble metal formulations operate in a PEM electrolyser at 0.3 A cm^{-2} whereas noble metal formulations operate at I >3 A cm^{-2} at 1.8 V, the latter are still favoured.

An efficient recombination catalyst based on a nanosized 30% Pt/TiO_2 was developed and produced on a proper scale. It was fully characterised in terms of structure, morphology and catalytic activity. Its use was more efficient down-stream the anode compartment resulting in H_2 content in O_2 stream at ppm levels (MS6).

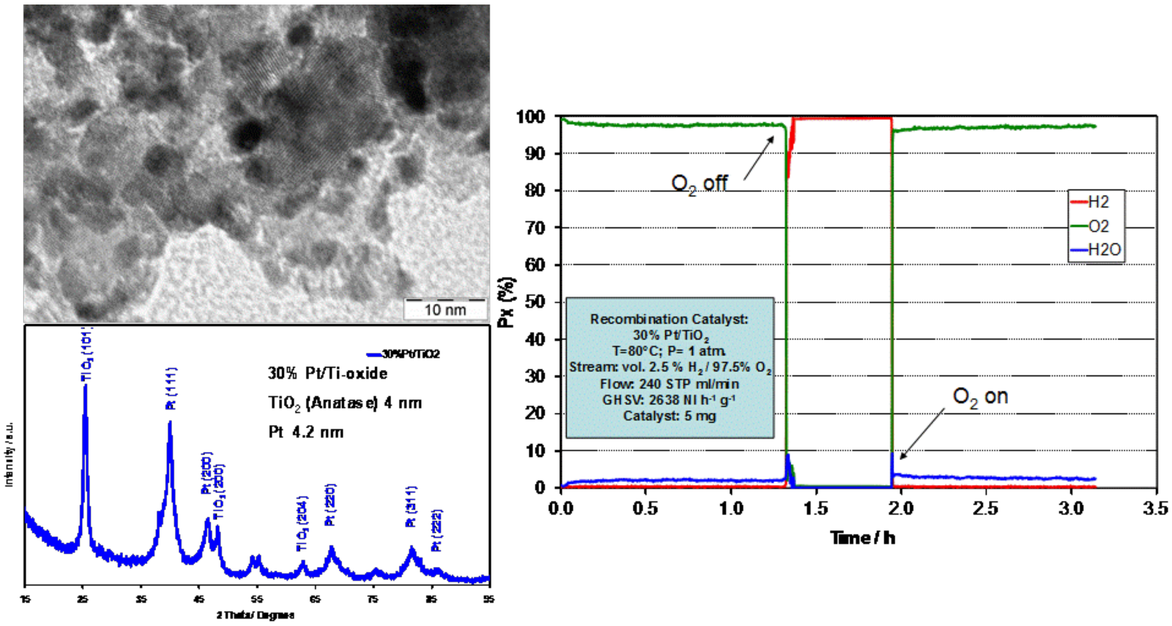
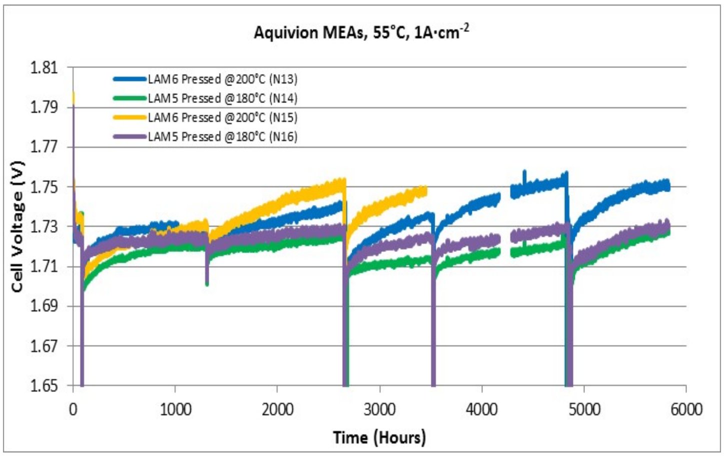


Fig. 14 TEM and XRD analyses of the Pt/Ti-oxide recombination catalyst and catalytic activity for hydrogen recombination down-stream the anode compartment (CNR-ITAE)

V. WP5 VALIDATION OF MEMBRANES AND CATALYSTS IN MEAS - WPL: ITM

The activities carried out in WP5 concerned with a validation in terms of performance and stability of the novel membranes developed in WP3 in combination with the enhanced catalysts developed in WP4 in practical MEAs with conventional and reduced noble metal loadings.

Several tests of almost 6000 hrs were carried out at ITM with MEAs based on the components developed in the project. Initial MEAs, regardless of their hot pressing temperature, showed a reversible degradation. CCMs pressed at 180°C showed lower voltage rise than those pressed at 200°C. The most stable MEAs gave a decay rate of 3.3 µV/h at 55 °C which was significantly smaller than the maximum limit fixed in the project i.e. 15 µV/h (MS9). The performance measured at ITM for these MEAs increased significantly with the increase of temperature. Performances were in line with project milestones (MS7).



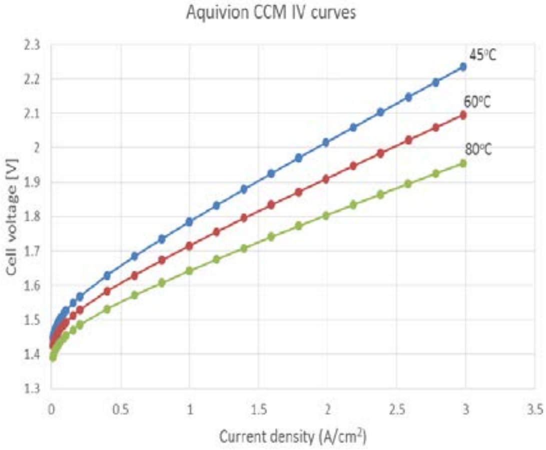


Fig. 15 Constant current tests at ITM of CCMs with different hot pressing conditions. Polarization curves of an Aquivion CCM in a lab scale cell at various temperatures (ITM).

The MEA behavior as function of temperature was also studied at CNR-ITAE. The best MEA performances were achieved for a catalyst prepared using a small batch procedure. At 90 °C, 1.6 V and 1.8 V, the current density values were 1.2 A cm^{-2} and 3.4 A cm^{-2}, respectively. The performances recorded at CNR-ITAE for small batch was not significantly different than for the large batch catalysts.

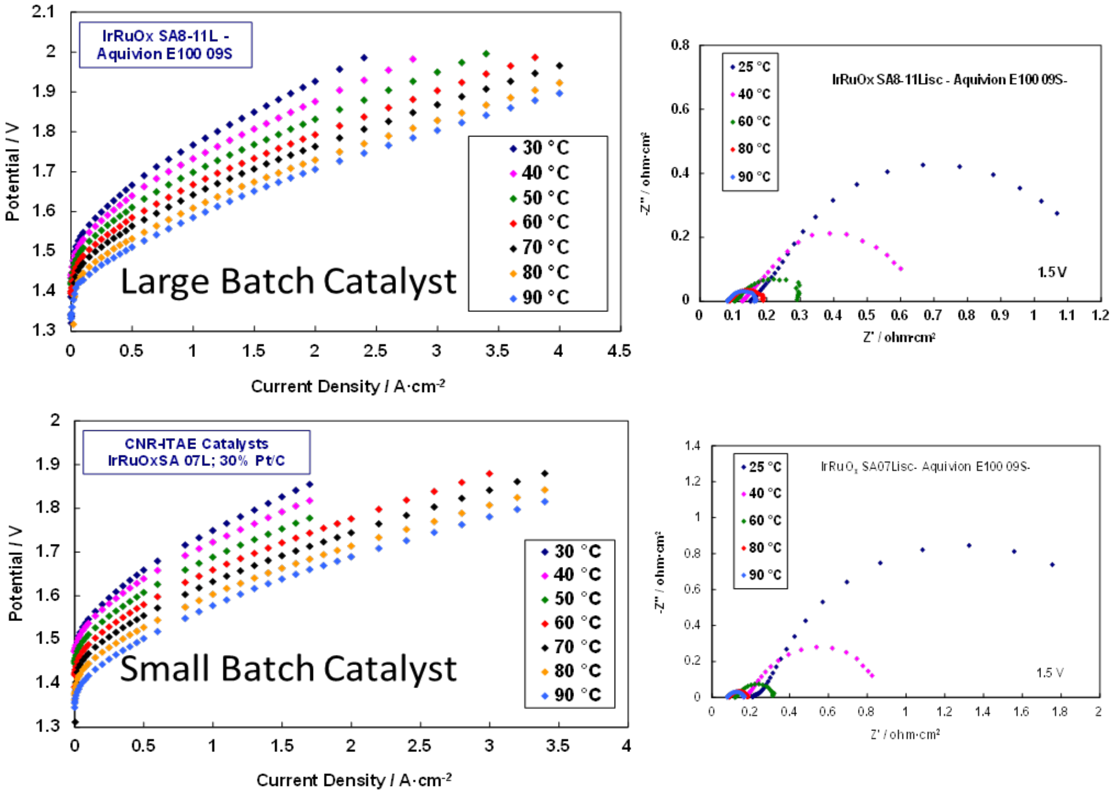


Fig. 16 MEA behavior as function of temperature: CNR-ITAE Large vs Small batch Ir$_{0.7}$Ru$_{0.3}$Ox –Pt/C catalysts with Aquivion E100-09S membrane

CNRS' focused into the performance study of water electrolysis at temperatures up to 140°C, using catalysts and membranes made in their laboratories, supplied by project partners and compared with reference materials. Performance studies using polysulfone reinforced membranes, based on different Aquivion ionomers with a range of equivalent weight, from 700 to 950 were performed. All these membranes were tested using a load of IrRuOx catalyst of 1.5 mg/cm^2 including a 33 wt. % of Nafion as binder for the anode and 33 wt.% for the cathode with a load of 0.5 mg/cm^2. At 80°C these MEA's, with an average membrane thickness of 35 µm presents a good performance in water electrolysis at 80°C. Compared with the Nafion based counterpart (Nafion N212), the global performance of these membranes was better than the reference at 80°C and extended the operating temperature range to 140 °C.

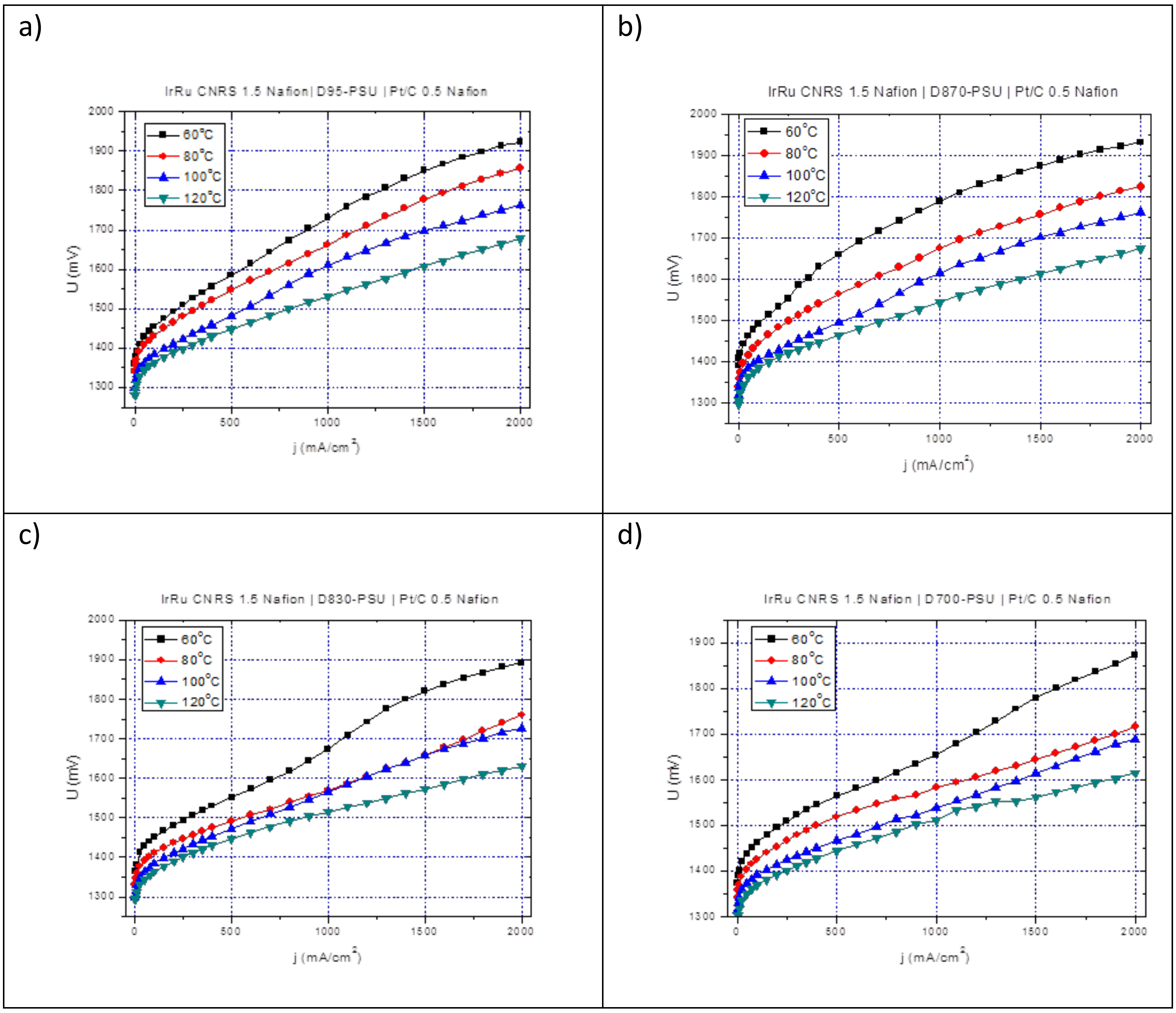


Fig. 17 PSU reinforced-Aquivion membranes performance tests at CNRS.

JRC's activity on WP5 was focused in the first phase on screening the MEAs provided by CNR-ITAE and, in the second phase, on the assessment of performance and stability of MEAs prepared with down-selected components at CNR-ITAE. Performances recorded at JRC with commercial MEAs were lower than those obtained using the MEAs developed in the consortium, based on Aquivion membrane and using CNR-ITAE catalysts, at similar noble metal loadings. The achievement of high performances and stability for the MEAs developed in the consortium is attributed to the enhanced proton conductivity and good mechanical stability of the novel membranes, the high electrochemically active surface area electrocatalysts, the optimized structural properties in terms of solid solutions and the segregation (enrichment) of Ir species on the surface for the anodic IrRuOx formulations.

Appropriate stability was observed in a durability test of about 500 hrs at JRC for the down-selected MEAs with no observable degradation at high current density. Just a very small performance decay at low current density and in the ac-impedance spectra was observed. This was in line with the project target. An interesting behavior noted is that the interruption of the test seems to "regenerate" the MEA that shows better performance at the restart of the test.

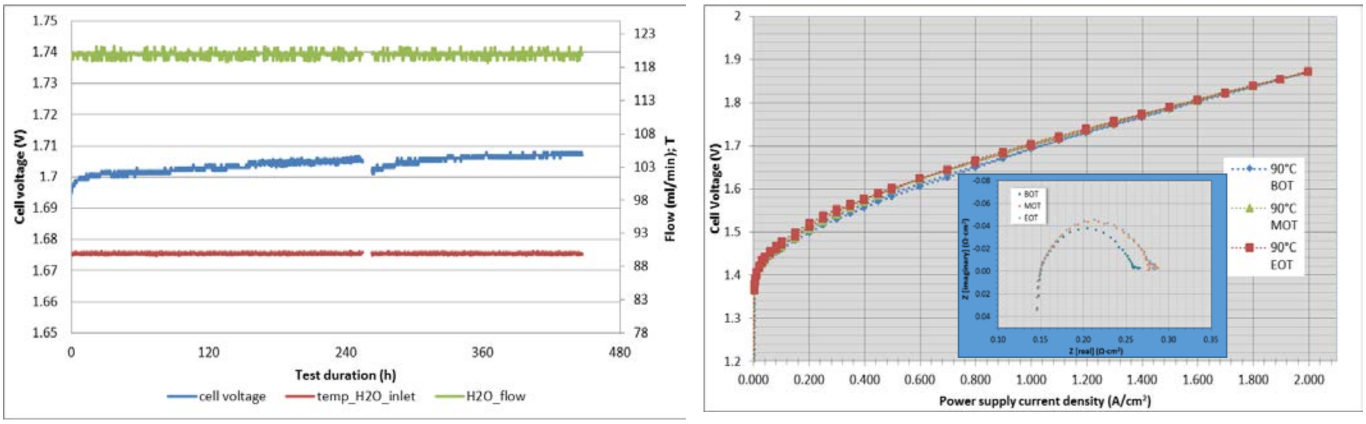


Fig. 18: IrRuOxSA03L - CNR-ITAE - Aquivion steady state degradation test at 1A/cm^2 at 90°C carried out at JRC. Polarsation curves and ac-impedance tests before and after the durability test (JRC).

In the second phase of the project, the activities carried out in WP5, were especially concerning with the manufacturing of large area MEAs (130 cm^2 active area) at ITM based on the scaled-up membranes and catalysts delivered from WP3 and WP4. ITM received Aquivion membranes (E098-09S) from Solvay and catalyst from CNR-ITAE (IrRuOx Black (SA8-11) and Pt/C (30 wt%)). ITM made catalyst inks using the catalyst from CNR-ITAE and sprayed them onto a large surface area membrane (130 cm^2 active area). Finished CCMs were hot pressed and hydrated for further testing.

In particular, large area MEAs at ITM were manufactured for both short and full stacks. Validation of performance and stability of the down-selected large area membranes in combination with the large batch catalysts in practical MEAs, with conventional and reduced noble metal loading under practical operation, was carried out (ITM). MEAs composed of CNR-ITAE nanosized Pt/C cathode and solid solutions IrRuOx anode catalysts, and Aquivion short-side chain perfluorosulphonic ionomer membranes have provided performances exceeding 3 A·cm^{-2} at 1.8 V terminal cell voltage at 80°-90 °C with hydrogen cross-over lower than 1 mA·cm^{-2} equivalent current density at ambient pressure. The achieved performance represents a 50% improvement over the project milestone (MS7) for MEA performance. This excellent performance was also approached using a total noble metal loading per MEA active area of less than 0.5 mg cm^{-2} (MS8). Both ITM and CNR-ITAE have shown stable performance for MEAs based on Aquivion membranes equipped with conventional (> 1200 hrs, 8 µV/h) and low noble metal loading. In particular, CNR-ITAE carried out durability tests (> 1000 hrs), for MEAs with different total noble metal loadings. The test with a catalyst loading of 1.5 mg cm^{-2} gave a cell voltage variation ~8 µV/h; this was better than the project target (MS9, 15 µV/h). Appropriate stability (cell voltage change ~15 µV/h) was also achieved for the low loading, 0.5 mg cm^{-2} MEAs tested at CNR-ITAE; this was in the range of the project target (MS9).

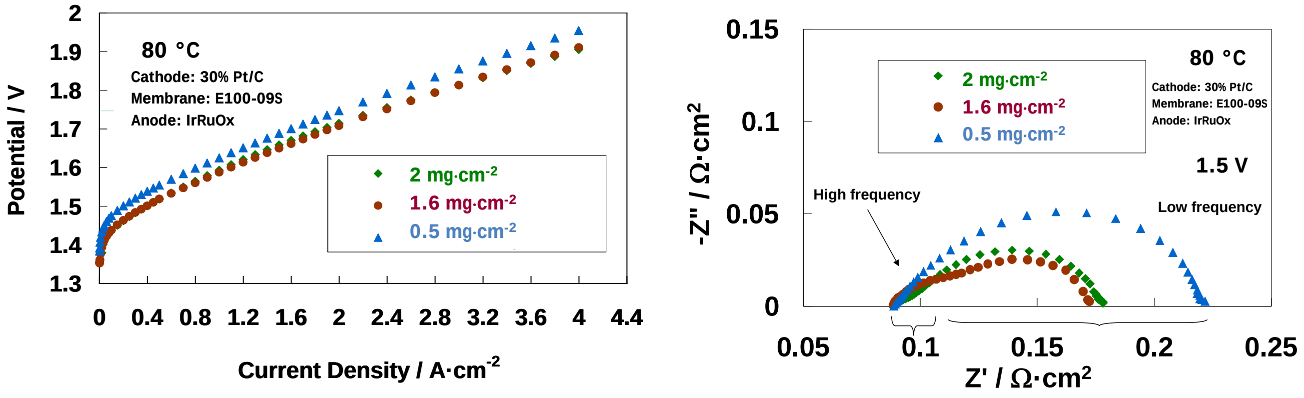
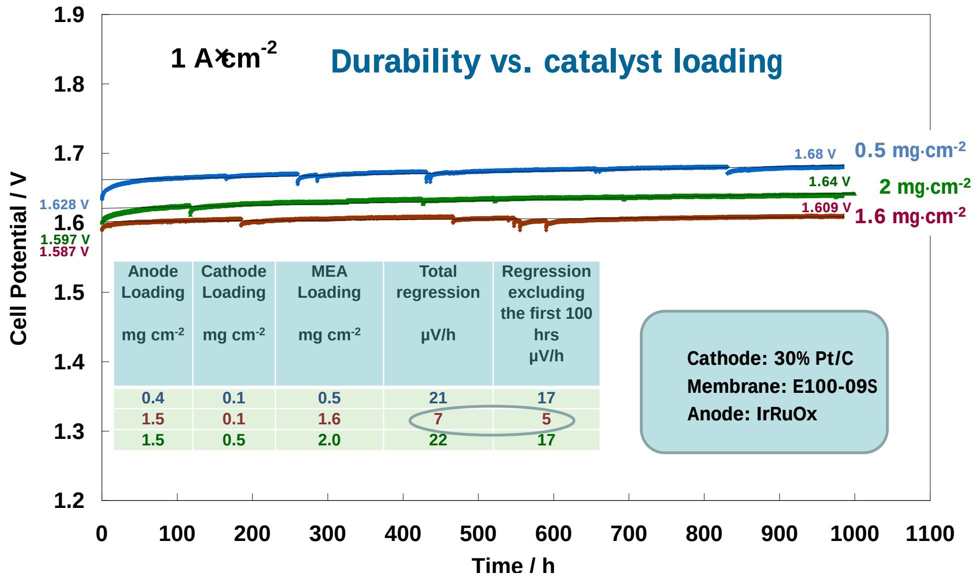


Fig. 19 Comparison of MEAs performance at 80 °C using different catalyst loadings and corresponding ac-impedance spectra at 80 °C and 1.5 V (CNR-ITAE)



Anode Loading mg cm-2	Cathode Loading mg cm-2	MEA Loading mg cm-2	Total regression µV/h	Regression excluding the first 100 hrs µV/h
0.4	0.1	0.5	21	17
1.5	0.1	1.6	7	5
1.5	0.5	2.0	22	17

Fig. 20 Durability studies at CNR-ITAE for MEAs with different catalyst loadings

ITM have scaled up the materials through their MEA production line. They have made catalyst inks using Aquivion(TM) dissolved ionomer, and measured the rheology of these. The particle size distribution of the catalyst was as expected with the Aquivion(TM) ionomer. Accelerated settling tests were performed on the inks to ensure the catalyst does not settle out in the lines. The results showed no difference to standard formulations. ITM have coated Aquivion membranes with catalysts to make production scale CCM's.

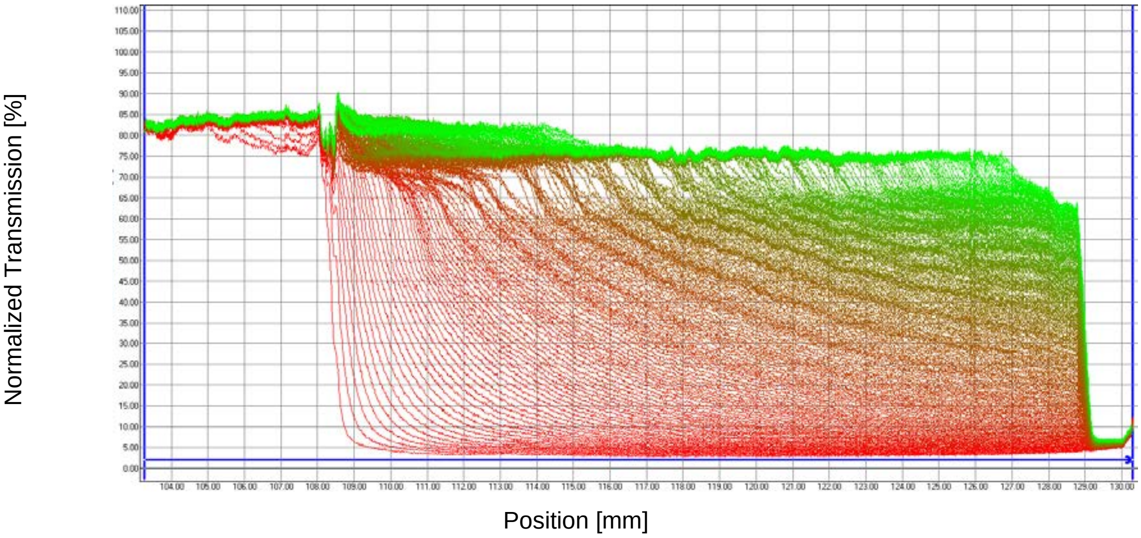


Fig. 21 The transmission gradient moving from red to green where the red line = T_0 moves to the green line T_n.(equivalent to one month) (ITM)

VI. WP6 STACK AND SYSTEM DEVELOPMENT AND TESTING - WPL: TRE

Non-active components of the stack and specific stack architectures have been individuated at ITM in the first phase of the project. The inside of the electrolyser stack has been reconfigured to eliminate expensive machining costs. The traditional “bipolar plates” have been replaced for lower costs components and injection moulded parts. High pressure (up to 30 bar) differential mode operation (pressurised hydrogen, unpressurised anode) was the preferred option. Preliminary characterisation of a small capacity water electrolysis systems was carried out in the first phase. An extensive stack testing was carried out in the second phase and allowed to assessing the validity of the newly developed polymer electrolyte membrane water electrolyser (PEMWE) components (membranes, catalysts and MEAs). MEA materials developed within the ElectroHyPEM project were tested in a production sized stack at ITM. Initially the materials were tested in short three-cell stacks in a stack testing apparatus designed and built for the ElectroHyPEM project.

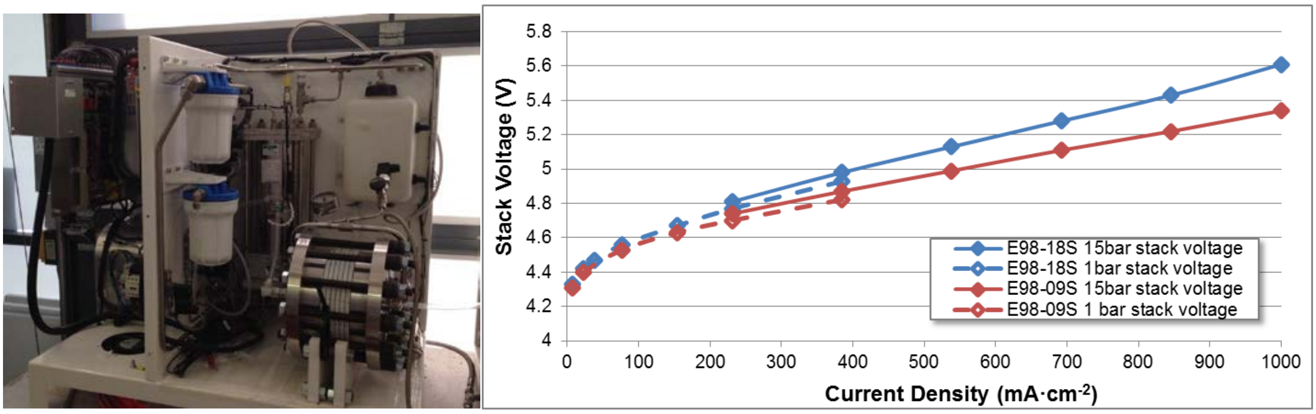


Fig. 22: Stack test rig built for ElectroHyPEM at ITM and short-stack testing. Stack polarization curves of 90 and 180 µm thick CCMs (ITM)

The thinner CCM made with 90µm Aquivion® membrane was selected for further testing. Polarization curves at different temperatures were recorded. As expected, increasing the temperature enhanced the kinetics and gave better performing CCMs. Cell voltages at 1A/cm2 for 50°C and 80°C were 1.78 V and 1.60 V, respectively. This is equivalent to a stack efficiency of greater than 92% vs. the higher heating value (HHV) of hydrogen.

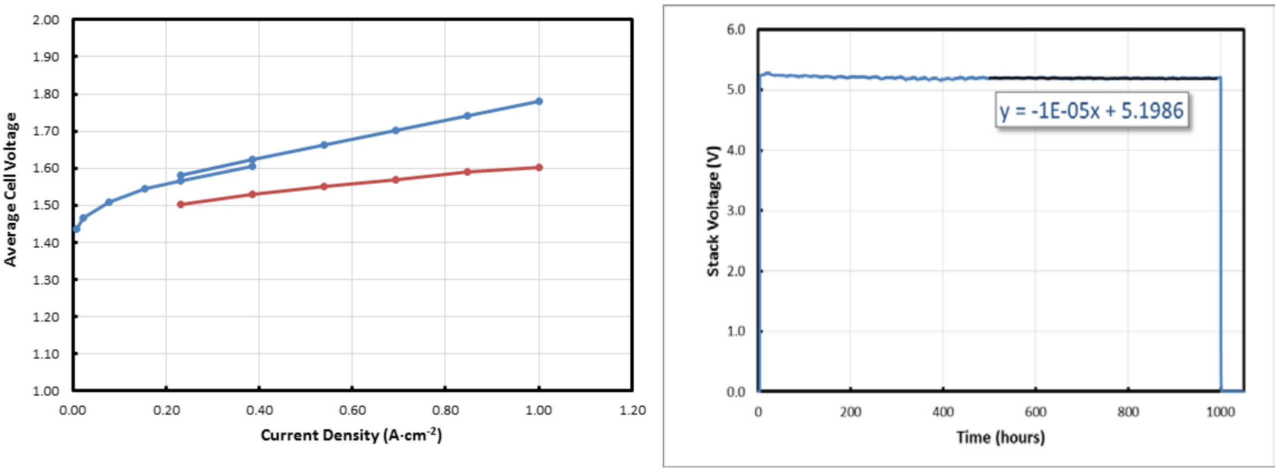


Fig. 21: a) Short-stack polarization curves of 90 µm thick CCMs (130 cm^2 active area) at 50°C (blue line) and 80°C (orange line); b) Stack voltages as a function of time at 1A/cm^2, 50°C. (ITM)

After initial polarization curves, stack went into a longevity test where current was held at 1 A/cm^2 and stack voltage was monitored at 50°C and 15 bar differential pressure. No visible degradation was observed during the 1000hr test. In addition to constant current testing, ITM also performed current cycling tests in order to mimic real system operation in the field. Stack was ramped up from 0 to 100% in 3 seconds. It was hold at 100% for 10 minutes and then descended from 100 to 0% in 3 seconds. It was hold at 0% for 1 minute. A minimum of 15000 cycles were applied to stacks to investigate the stability and also understand system degradation.

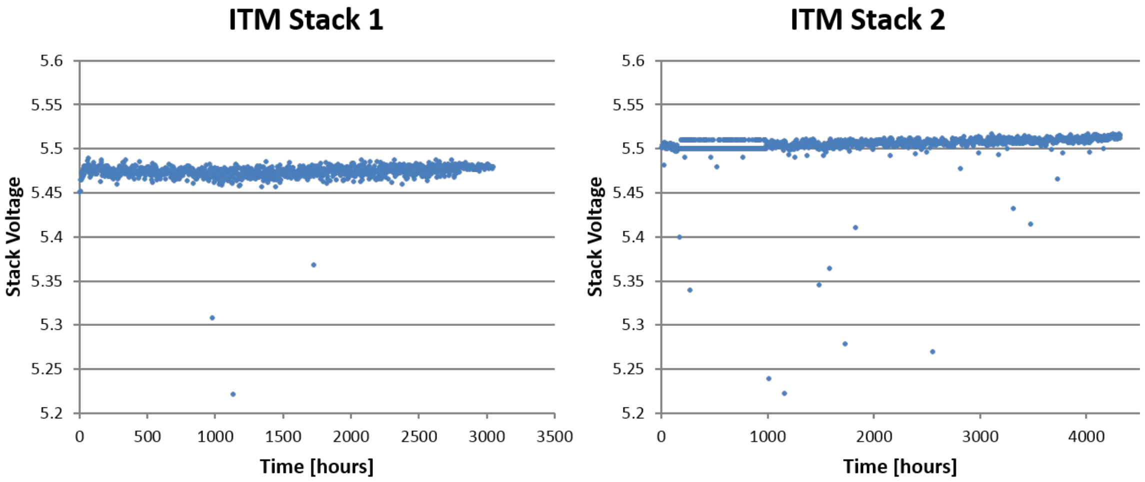


Fig. 24: Power cycling data of two short stacks at ITM

It was observed that the current cycling does not cause measurable degradation during the period of extensive testing on large scale short stacks.

To interface the electrolyser stack to a renewable source, it is important that the stack is able to respond quickly from off (cold start) as well as from on. An investigation into the response time of the PEM electrolyser stack was made by acquiring information about the rate in response to fluctuating power profiles. A controlled current step was applied to the stack, whilst the voltage of each cell is monitored at a frequency of 5000 Hz. To understand the response of the system to rapid changes in current when operational (such as might be seen when the system is connected to an intermittant power source such as wind or solar), the system was altered from 90 A (full power) to 45 A (half power) and then back to 90 A. The response of the electrolysis system was very fast, with the stack reaching its working voltage in significantly less than 0.1 seconds.

Nearing completion of the project the final set of materials for stack demonstration and testing was agreed upon by the project partners. This consisted of a CCM utilizing Aquivion® membranes (E098-09S) from Solvay and catalysts from CNR-ITAE (IrRuOx Black (SA8-11) and Pt/C, 30 wt%). The catalyst inks were made at ITM and the rheology tested prior to manufacture. The catalyst inks were ultrasonically sprayed onto a large surface area membrane (130 cm^2 active area) using a robot arm to ensure an even coating. Finished CCMs were hot pressed and hydrated following the conditions developed earlier in the project for working with Aquivion® membrane and ionomer.

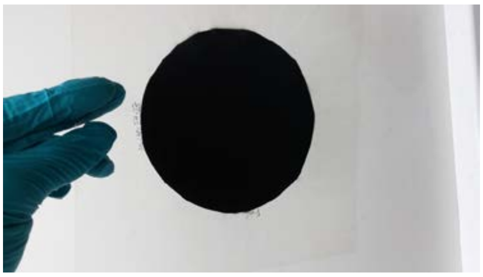


Fig. 25: Large area (130 cm^2) CCM made at ITM using down selected project materials (Solvay membrane and CNR-ITAE catalyst)

The materials were made into a 20 cell stack with a total surface area of 2600 cm^2. The stack materials were tested for durability under a different load conditions including:

1. Beginning of test (BoT) and end of test (EoT) characterization; including polarization curves at different temperatures and impedance measurements
2. Steady-state testing for 1000 hours at 1 $A\cdot cm^{-2}$ (55°C, 1.2 Nm^3 hydrogen per hour)
3. Load profile testing mimicking wind profiles
4. ITM-developed AST

Fig. 26: 20 cell stack made and tested using down selected materials from partners. (ITM)

The stack produced 1.2 $Nm^3\cdot h^{-1}$ of hydrogen (WP6) with an energy consumption ~3.5 kWh/Nm^3 H_2; this was better than the project target (MS10). The stack performance was measured at different temperatures and run at full operation for 1000 hours showing very little degradation (< 10 $\mu V\cdot h^{-1}$) (MS11).

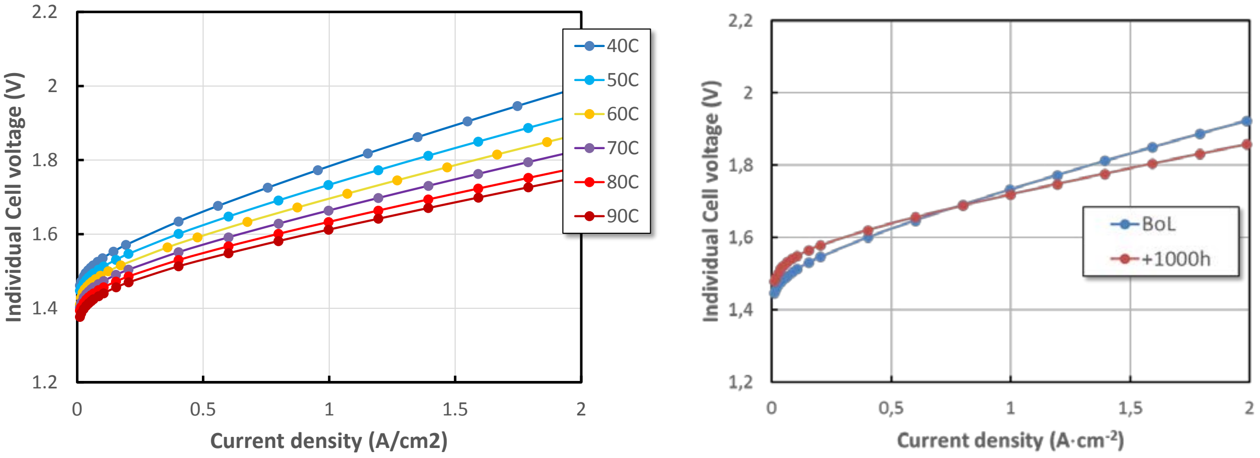


Fig. 27: Beginning of life IV curves for the Aquivion® stack CCM (ambient pressure). IV curves for the final stack at beginning of life (BoL) and after 1000 hours of operation at 1A·cm^{-2} (55°C, ambient pressure) (ITM).

A micro wind turbine (7 kW) for residential use was developed at TRE for renewable energy source (RES) coupling evaluation of the PEM electrolyser. This was full assessed for one year and the power-time characteristics were provided to ITM for stack testing under conditions simulating RES coupling.

The main characteristics of the selected wind turbine prototype were: Rated power 7 kW at a wind speed of 13 m/s; assisted cut-in at wind speed of 4.5 m/s; cut-out at wind speed of 20 m/s.

Fig. 28: 7 kW TRE wind turbine installed in a remote location.

The electrolyser stack was thus tested using a model operating cycle based on real wind data collected within the project (MS12). It showed very little degradation (<8 µV after 10 000 cycles). The stack was also tested against an accelerated test protocol developed within the project based on cold start (WP6).

The real wind profile for a typical 24 hour period (taken at random) has been converted to the current load that the electrolyser would be expected to absorb. It can clearly be seen that steady state operation only provides part of commercially relevant testing. Field operation is highly irregular and places different strains on the MEA than the constant current tests. Unpredictable operation needs to be modelled in the lab via a simulated load profile to accelerate the stress seen by the electrolyser.

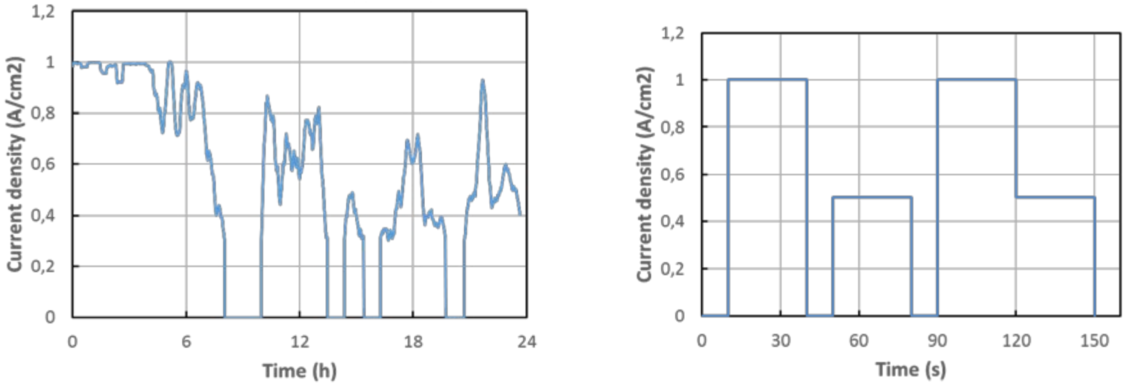


Fig. 29: Variable current load the electrolyser would be expected to handle over a typical 24 hour period. Model used to simulate the real load (ITM).

The MEAs inside the stack were stressed via this load profile to 10 000 start-stops, which is equivalent to several years intermittent operation. The voltage of the stack was monitored as this is the key performance indicator for stack degradation.

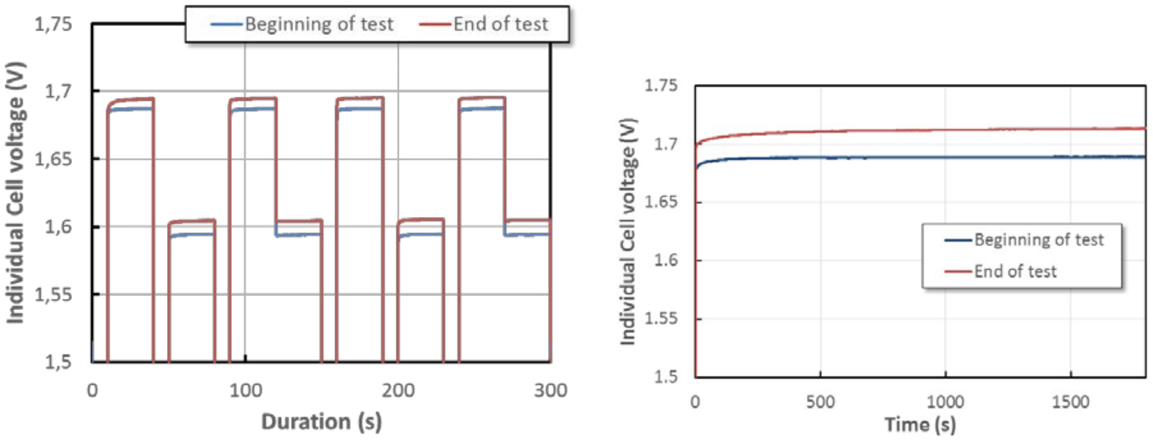


Fig. 30. Voltage profile of an individual cell in the ElectroHyPEM stack before and after 10 000 cycles. ElectroHyPEM stack voltage (constant current measurement; single cell shown) before and after voltage cycling in the AST. (ITM)

The MEA's responded well to the load cycling test, showing a very small 8 mV rise in cell voltage after 10000 current cycles. It is clear that rapid changes in the current density and rapid on-off occurrences do not significantly damage the MEA. From these results it is derived that MS12 is achieved: Assessment through coupling of PEM electrolyser with RES or testing under simulated RES current profiles.

A voltage-controlled accelerated stress test was performed on the ElectroHyPEM final stack. The stack was cycled 1000 times between 1.75 V and 0 V, which is representative of roughly 3 years of in-field intermittent operation. The Aquivion CCM's responded well to this test, gaining roughly only 25 mV of voltage rise during the test. This voltage rise is indicative of the stability of the catalysts, and it shows that the Aquivion® CCM's and stack would be expected to last for several years of intermittent operation.

The project was ultimately successful delivering a new commercially viable MEA that has been testing in a production sized stack. Further work in ongoing to commercially exploit the technology.

VII. WP7 DISSEMINATION, OUTREACH, EXPLOITATION - WPL: CNRS

Regarding the dissemination and exploitation activities, the main achievements for the second period concerned mainly on dissemination of project information achieved through various ways depending on the audience:

- Updating the dedicated project website (http://www.electrohypem.eu)
- Journal publications (7 published articles on international peer-reviewed journals achieved)
- Conference presentations (15 contributions achieved with 2 invited)
- Participation of Electrohypem partners to the SUSGHEN workshop in Paris (3 contributions) and Electrohypem workshop on PEM electrolysis in Taormina (7 contributions) with project results presentation.
- Public awareness: information activities to increase public awareness of hydrogen generation through water electrolysis during dissemination activities addressed to university and high school students with the visit to the research laboratories, etc.

The coordinator has dissemination the project results by representing the project at international conferences, in particular with two oral presentations at the FCH-JU review days 2012 and 2014 and poster presentations (2013, 2014).

Exploitation, dissemination and training of young researchers were also relevant aspects of the project.

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

VIII. POTENTIAL IMPACT

The Electrohypem project has specifically concerned with research on functional materials and technology for PEM electrolysers as well as their coupling to renewable energy sources. The expected impact of the project is related to the sustainable hydrogen production which can meet an increasing share of the hydrogen demand for energy applications from carbon-free or lean energy sources. R&D in innovative hydrogen production from renewable energy sources is reported among the priority topics for this application area. The scope of the project was to carry materials research to reduce the total life cycle costs related to the present PEM electrolysers by replacing current commercial materials for membranes, catalysts, bipolar plates with lower cost materials.

The main disadvantage of PEM electrolysis is at the moment the high cost, which is mainly due to the use of noble metal catalysts, perfluorinated membranes and titanium current collectors. In order to be competitive within the field of decentralised hydrogen production and especially for small scale applications under operation with renewable power sources, the PEM electrolyser must be reasonably cheap; moreover, it should be compact, characterised by high durability and capable of producing hydrogen at high efficiency and at suitable pressure. At present, challenging problems to the development of such systems remain. Electrochemical water splitting is associated with substantial energy loss while materials degradation and high cost represent relevant issues.

All these aspects have been addressed in the project and specific quantitative project milestones have been achieved. In particular electrolysis performances > 3 A cm^{-2} at 1.8 V terminal cell voltage achieved in this project using novel and cost-effective materials represents a substantial reduction in capital costs. These are inversely related to the operating current density. Some concern is related to performance stability since highly conducting membranes and high electrochemical surface area electro-catalysts in general show lower stability. This evidence has required in the project the introduction of novel solutions to enhance stability while keeping the performance at proper level and a compromise was adopted between the different material properties to obtain proper combination of performance and durability.

Achievement of a degradation rate <10 µV/h and stacks with energy consumption < 4 kWh/Nm^3 H_2 represent a suitable progress beyond the state of the art. Coupling with renewable energy sources such as micro wind turbine for residential uses can allow for a wide scale application of these systems. A cost analysis was made within the project and significant advancements towards the reduction of capital costs have been identified.

The active components currently contribute to about 70% of the stack cost whereas the BoP contributes by about 20% of the overall systems costs. Thus, it is evident that a considerable cost saving may be achieved by using stable, efficient and cost-effective materials. Innovative membrane, low noble metal loading electrocatalysts and cost-effective bipolar plates developed in the project can thus significantly contribute to reduce the present costs of PEM electrolyser components. The cost analysis has indicated good perspectives to achieve stack capital cost (CAPEX) <2.5 kEuro/Nm^3 H_2/h.

Due to the significant cost reduction associated with the production process of the Solvay Aquivion ionomer, the utilization of noble metals at low loadings with novel MEA forming procedures, the innovative cost-effective stack design etc., a suitable perspective for mass production is assessed. As derived from an internal cost analysis (D6.2), the combination of CCM savings and coatings developed in ElectroHyPEM can lower the cost of a production stack by 26% and the increase of the operating current density can also reduce the CAPEX considerably.

These advanced electrolyser systems can be matched directly with high coupling efficiency to the small wind turbines (7-10 kW) and find suitable application for residential uses and remote generation.

The progress beyond the state of the art achieved in the project is outlined below:

- Performances > 3 A cm^{-2} at 1.8 V vs. 1 A cm^{-2} at 1.8 V cell voltage at 80 °C.
- Total MEA noble metal loading 0.5 mg cm^{-2} vs. 3 mg cm^{-2}.
- Stability improvement ~8 µV/h at 1 A cm^{-2} vs. 20-30 µV/h in durability tests of 1000 hrs.
- Operating temperatures up to 140 °C vs. 55-80 °C with performance of 1.5 V at 1 A cm^{-2} at 140 °C.
- Stack Energy consumption ~ 3.5 kWh/Nm^3 H_2 at current density of 1 A cm^{-2} vs. 4.8-5.6 kWh/Nm^3 H_2 state-of-the-art.
- From 2 to 4 times reduction in the cost of MEA components (catalyst and membrane) through the reduction of noble metal loading and the use of cost-effective membranes.
- Significant progress in the developments of cost-effective bipolar plates

These results may effectively contribute to the road-map related to the achievement of a large-scale decentralised hydrogen production infrastructure and the next step is the scaling up of the products and devices developed in the project.

IX. EXPLOITATION

Scalability, reliability and cost-savings are relevant aspects for the deployment of PEM electrolysis devices. These features were specifically addressed during the Electrohypem project and are fully described in the report D7.3. The industrial scalability of both down-selected components and fabrication processes was assessed in particular for membranes and electrolysis stacks. The down-selected PEM electrolyser components developed in Electrohypem were analysed in terms of performance and cost, and have been demonstrated in a 4 kW system. The dynamic behaviour has been validated for coupling with mini wind turbines. The performance and stability targets obtained in the project represent significant progress to achieve a good reliability for the PEM electrolysis devices. These innovative components and solutions for the PEM electrolyser were analysed in terms of industrial scaling-up. Estimation of the costs for in series production, carried out in D6.2 indicates that significant cost savings for the PEM electrolysis technology are possible by integrating the novel solutions developed in Electrohypem. Scaling-up programmes for specific components, e.g. the Aquivion membranes, have already started and exploitation plans have been addressed.

The core business of the industrial partners (Solvay, TRE, ITM) involved in ElectroHyPEM project and their plan for exploitation is briefly described in the following.
Solvay is currently commercializing ionomer membranes for fuel cells but not a specific product for the PEM water electrolysis application; since the interest in electrolysis is growing, the natural exploitation of the results of the project is the creation of a line of membranes dedicated specifically to this application. TRE is active in the field of renewable energy sources. The results of the project will be used to implement renewable power sources with cost-competitive electrolysis plants for different applications and especially as a means of storage of surplus energy. ITM is presently producing a set of commercial PEM electrolysers; project results will be exploited to implement current products and expand the range of applications.

Thus, the consortium covers the manufacturers of basic materials, MEA, stack, system for PEM electrolyser as well as an end-user developing and commercialising renewable power sources including wind plants. Exploitation and use of the project results are essentially addressed by the industrial partners. A narrow collaboration between research organizations and industry makes possible a rapid exploitation of the new components and their integration into practical systems. Key-issues such as patents, licenses, royalties and dissemination of the intellectual property arising from the project, and coordination activities related to negotiations between the Consortium and external parties are dealt by the project steering committee. Several promising materials showing enhanced performance with respect to the corresponding benchmarks are actively evaluated by the industrial partners and are taken under consideration for their future system developments. Activities related to the analysis of interfacing PEM electrolysers to renewable power sources such as small wind turbines have brought significant new knowledge that can be the subject of exploitation within consortium.

The fact that this project team comprises a strong participation of companies with established development, manufacturing and commercial activities, facilitate the exploitation of the results, since the industrial partners can immediately explore any new successful development. In general, the industrial partners have significant experience on exploitation of the components related to this technology. Thus, the successful development of new products in the Electrohypem project will add to the value of their product portfolios. According to the Electrohypem Consortium Agreement, the industrial partners can also seek to inform potential customers of positive results obtained in the project in accordance to the IPR considerations. Regarding the time-scale for the exploitation of the materials, components and devices developed within the project, this can be reasonably identified in a few years after project termination to properly assess them also in terms of scaling-up, cost-effectiveness and durability.

Specific exploitation plans of the industrial partner in Electrohypem are presented below:

Solvay Specialty Polymers is committed in developing material solutions for novel and green energy systems and aims to become a world leader of solutions for grid stabilization and developing ionomers for water electrolysis is part of this strategy. SOLVAY has a significant experience and knowledge on the production of Aquivion short-side-chain PFSA materials, currently commercialized for fuel cells.

At the start of the project, SOLVAY has produced and supplied partners with baseline materials for MEA assembly. Baseline membranes were extruded membranes based on the Aquivion ionomer with an equivalent weight of 870 g/eq, a type of ionomer firstly developed and commercialized for the specific application of fuel cell. The improved products developed throughout the Electrohypem project for water electrolysis focused on maintaining the same technology of manufacturing that is the extrusion, improving the mechanical consistency, reducing gas crossover and maintaining the conductivity unvaried and, reducing the cost. Increasing the equivalent weight of the material gave the desired effect since it increased the portion of the material that can crystallize. The increase in crystallinity allowed the production of membrane with high mechanical stability due to less swelling of the membrane, a critical issue when operating at high temperature water electrolysis. These improved polymers were melt-extruded in membranes with different thickness (50-90-180 micron). The equivalent weight of the ionomer is ~1000g/eq (roughly corresponds to one unit carrying the sulfonic group pendant every seven units of TFE), the polymer was chemically stabilized by treating the polymer powder precursor with fluorine gas so to remove the side chain end groups (chemical degradation resistance increased compared to standard ionomer). The membranes were converted from precursor form ($-SO_2F$) to the active one (-

SO_3H) via a treatment in KOH followed by an acid exchange in HNO_3, such treatment was done directly on the film.

The different type of extruded membranes with high equivalent weight have been subjected to extensive ex-situ and in-situ characterization by the different partners involved in the project. The collected data allowed SOLVAY to assess and compare the different membrane produced in terms of relevant parameters and to down select a promising membrane for scale up and production capabilities.

The selected E100-09S extruded PFSA achieved most milestones such as proton conductivity target of 0,2 S/cm at 95 °C and area specific resistance of 46 mOhm.cm^2. The developed grade was able also to match H_2 permeability targets at ambient pressure. Hydrogen crossover at 15 bars differential pressure and 55 °C was also checked on this grade and showed hydrogen concentration at anode lower than 2%. Moreover, E100-09S showed relevant performances more than 1000 hrs of durability at 55 °C and 80 °C at 1 A/cm^2.

The polymerization of EW=1000 g/eq ionomer has faster kinetic compared with the baseline material, since TFE is the most reactive monomer, and, considering also that TFE is cheaper than SFVE by far, the overall production cost of the E100-09S membrane is reduced in comparison to the E87-12S baseline membrane.

The process and the main production steps of fabrication of the said membrane to an amount sufficient for the MEA development in WP5 and beyond have been optimized to have a minimum number of unit operations with an efficient protocol for each phase. Criteria are, among other: (i) optimization of process yield and quality (ii) process efficiency (iii) minimization of by-products and scraps. After preliminary tests at pilot plant scale, the process has been validated also at industrial level. This allowed the creation of a production line of membranes dedicated specifically for this application. The production capability of this promising material is estimated to 10000 m^2 per year; Solvay will commercialize this developmental material in the next years. As a first step this promising ionomer will be included in the Solvay Catalogue of Products and specific agreement can also be established with potential partners with preference but not exclusive to the ITM project partner which is one of the EU leader for manufacturing PEM electrolysers.

Solvay Speciality Polymers website: . http://www.solvayplastics.com/sites/solvayplastics/EN/specialty_polymers/Pages/solvay_specialty_polymers.aspx

ITM Power are one the world's leading water electrolysis companies producing PEM water electrolysers for both energy storage and renewable transport applications. The core technology for ITM is the electrolyser stack, although ITM build and supply fully integrated "*plug and play*" systems including the balance of plant, water and hydrogen purification and the control and safety systems.

The EU's target for Renewables - in the new Climate and Energy Framework for 2030 one speaks of at least 27% RES for the EU (Germany speaks of a 50% target in 2030 and 80% in 2050) - will inevitably lead to high peaks of electricity due to the intermittency of solar, wind, etc. This trend will increase the more RES comes online. To meet the future challenges of an energy system based on renewables Europe needs a variety of reliable storage technologies that can give a rapid response to the intermittent renewables.

Water electrolysis is one such energy storage technology. It is simply an energy conversion system used to turn electrical energy into chemical energy, in this case hydrogen, thus providing a means to store the electrical energy. If the electrical energy comes from a renewable energy source then the hydrogen produced has a very low (or zero) carbon footprint (green hydrogen).

Whilst hydrogen technology has the ability to revolutionize societies reliance on fossil fuels, there must be a commercially viable business case to ensure widespread uptake of the technology. To ensure the future commercial viability of water electrolysers, there needs to be improvements in efficiency, capital cost, and lifetime. All three of these aspects were improved upon within this project.

The ElectroHyPEM project enabled ITM to work with the partners to produce a more efficient, lower cost, and longer lasting electrolyser stack. In addition the project allowed the ability for ITM Power to work with Tozzi Renewable Energy, a potential customer/end user to ensure the product met future customers' requirements.

Within the project, modifications to the catalyst manufacturing process were developed and lower catalyst loaded MEA's were manufactured and tested for durability against a model renewable energy source. Both of these developments been transferred from the laboratory into production and the first deployment of this technology will be in a 500 kW renewable energy storage product by early 2016. These developments, which wouldn't have been possible without the help of the European community and the FCH-JU, have already resulted in a significant cost saving to ITM's electrolyser stacks.

Stack improvements developed within this project are also being transferred into production, and it is envisaged that these will be used in products manufactured from early 2016 onwards. They have resulted in further cost savings. The combined cost saving are predicted to result in a 24% saving in stack costs.

Another result that came from this project was an increased stack durability. In a commercial product it was expected that over the lifetime of the product the stacks will need to be changed three times. The increased durability of the materials developed within this project have predicted that the stacks will only have to be changed once in a twenty year lifetime. This will result in a significantly lower maintenance cost.

ITM worked with Solvay to investigate putting new Aquivion membranes developed within this project into a product. The new MEAs developed in this project containing these membranes have been tested at commercial stack scale within the project. It is still early days but ITM are continuing to work with Solvay to fully qualify these materials in an electrolyser product.

This project has been extremely successful from ITM's point of view. It has brought together materials developers with manufacturers and potential customers. The early results from the project are already in production and will be on the market by early 2016. It is expected that the commercial impact of this project will continue for many years as the materials developed make it to market. In addition, it is expected that the commercial relationships developed with the project will last for many years into the future and ITM looks forward to a continued and fruitful commercial relationship with these partners.
ITM Power website: http://www.itm-power.com/

TRE Tozzi Renewable Energy is active in the development and management of solar plants, on shore & off shore wind farms and biomass (both solid and liquid) power plants through different subsidiaries. Through Tozzi Holding subsidiaries, TRE is also active as EPC contractor for renewable power plants. Through its subsidiary Tozzi Nord, TRE is commercializing mini-wind turbines with peak power from 7kW to 60kW. TRE is also involved in research and development projects related to the production of solar cells, small wind turbine generators, new energy storage solutions (electrochemical batteries and electrolysers) and all the components related to these businesses.

All sources of electrical power have some degree of unpredictability, and demand patterns routinely drive large swings in the amount of electricity that suppliers feed into the grid. Wherever possible, grid operations procedures are designed to match supply with demand at high levels of reliability, and the tools to influence supply and demand are well-developed. The introduction of large amounts of highly variable power generation, like the ones coming from renewables, may require changes to existing procedures and additional investments. The use of small amounts of intermittent power has little effect on grid operations. Using larger amounts of intermittent power may require upgrades or even a redesign of the grid infrastructure. When considering wind farms, at times of low or falling demand where wind output may be high or increasing, grid stability may require lowering the output of various generating sources or even increasing demand, possibly by using energy storage to time-shift output to times of higher demand.

Hydrogen fuel storage and fuel cells have the scope to counter the intermittency of certain renewable sources, such as wind turbines, which increases the dependability of the source. The main advantage of using renewable sources, such as wind, to power the electrolysis processes to produce pure hydrogen for fuel storage or fuel cells, over other fuel sources, is that the by-products of hydrogen storage are benign, producing heat and water, with the former offering potential renewable heat advantages, and the overall process has an extremely low carbon footprint.

As we have been able during ElectroHyPEM project to demonstrate good perspectives for an efficient and durable coupling of PEM electrolysers with mini-wind turbines (7-10 kW power), TRE company is planning to include electrolyser units for on-grid or off-grid energy production & storage configurations, expanding its commercial offer. This includes already at the time being energy storage systems based on electrochemical batteries exclusively.

TRE Tozzi Renewable Energy website: http://www.tre-energia.com/ricerca-sviluppo.php

X. DISSEMINATION

The main objective of the dissemination and outreach activities was to guarantee proper diffusion of knowledge and project results according to an agreed strategy aimed to secure maximum impact with respect to the Intellectual Property Rights.

Dissemination and outreach activities arising from the ELECTROHYPEM project have been carried out and are listed below:

- Research publications in peer-reviewed journals,
- Meetings, conferences,
- Website,
- Outreach activities
- International workshop

The consortium has attended prominent international conferences, workshops and symposia. All presentations have followed the dissemination protocol to protect IPR.

The Electrohypem website (http://www.electrohypem.eu/) was designed to fulfil project communication and dissemination needs of the project towards the scientific community and the public by providing information including:

- project overall objectives, partner & work packages information

- public deliverables e.g. on specifications and protocols for assessing PEM electrolysis components and devices
- project activities: news, meetings, publications
- project resources: links, related events …
- project contact information

The project website has been updated regularly with information on project activities (news, meetings, publications, public deliverables ….) and project resources (links, related events etc.). The website will be kept as a source of information source of the activities performed in the project, and will also continue to receive and publish information on future published papers from the project.

Dissemination protocol

To respect the confidentiality of partners' results, and intellectual property rights, a dissemination protocol was elaborated and agreed. This protocol has been and will continue to be used each time a partner wants to disseminate results obtained within the context of Electrohypem.

LIST OF CONFERENCE PRESENTATIONS

The consortium has attended prominent international conferences to present results orally and in poster form made on behalf of the consortium. The complete list is provided in the report D7.2, and on the project website.

1. Investigation of Short Stack PEM Electrolyzer Based on nanosized Electrocatalysts
   A. S. Aricò, S. Siracusano, V. Baglio, N. Briguglio, A. Stassi, A. Di Blasi, R. Ornelas, V. Antonucci.
   GEIERA2012 (Italian Division of Electrochemistry), ENE-O-15, Abstract at p. 62, Libro degli Atti di Convegno GEIERA2012, M. Lo Faro, Vincenzo Baglio

2. Investigation of Baseline and Innovative Components for a PEM Electrolyzer in the Framework of the FCH-JU ELECTROHYPEM Project
   Plenary lecture: - A. S. Aricò
   EmHyTec 2012 - Euro Mediterranean Hydrogen Technologies Conference
   Book of Abstracts pp. 3-5 Conference Proceedings

3. The development of novel super powerful PEM electrolyser membranes,
   D. A. Greenhalgh, R. M. Lister, N. van Dijk, K. A. Yeomans,
   Electrochem 2013, Southampton, September 2013

4. Sulfonated Polysulfone-based Membranes for PEM Electrolyzer Applications S. Siracusano,V. Baglio,A. Aricò, O. Di Blasi, P. Staiti, F. Lufrano International Society of Electrochemistry Topical Meeting, Pretoria, South Africa, April 2013

5. Developments of components for a PEM Electrolyser
   E. Modica, S. Siracusano, N. Briguglio, A. Stassi, C. Alegre, G. Brunaccini, R. Bonelli, F. Matteucci, V. Antonucci, A. S. Aricò,
   Zing Conference, California, July 2013

6. Investigation of Ti-based supports for PEM Electrolyzer Applications
   S. Siracusano, A. Stassi, E. Modica V. Baglio, A. Aricò,
   Hypothesis 10th Anniversary, Edinburgh, July 2013

7. Investigation of a PEM Water Electrolyzer Based on a Sulfonated Polysulfone Membrane.
   S. Siracusano, V. Baglio, A. Aricò, O. Di Blasi, P. Staiti, F. Lufrano
   224th ECS Meeting, San Francisco, California, October 27 – November 1, 2013

8. Characterization of IrO2 and IrRuO2 Electrocatalysts for the O2 Evolution Reaction in PEM Water Electrolyzers.
   S. Siracusano, V. Baglio, C. Alegre, A.S. Aricò
   European Hydrogen Energy Conference (EHEC) 2014. March 2014 - Seville, Spain.

9. Development of composite membranes for medium temperature PEM water electrolysis using short-side-chain Aquivion,
   S. Giancola, A. Skulimowska, A. Reyes-Carmona, M. Dupont, S. Subianto, S. Cavaliere, D. Jones, J. Rozière, E. Moukheiber and L. Merlo
   Fuel Cells 2014 A Grove Fuel Cell Event, April 2014, Amsterdam

10. Aquivion® PFSA membrane for solid polymer electrolyte electrolyser applications.
    S. Siracusano, V. Baglio, L. Merlo, E. Moukheiber, A. S. Aricò.
    International Conference on Clean Energy (ICCE) 2014. June 2014, Istanbul.

11. Sustainable hydrogen production by PEM electrolysis for e-mobility.
    Jose Manuel Subiñas Seco de Herrera.
    International Conference on Clean Energy (ICCE) 2014. June 2014, Istanbul.

12. Performance Comparison between Iridium Oxide and Iridium - Ruthenium Oxide as Electro-Catalysts for PEM Electrolyzers
    C. Alegre, S. Siracusano, V. Baglio, A.S. Aricò
    225th ECS Meeting. May 11-15, 2014 – Orlando, FL.

13. PSU Nanofibre Reinforced Aquivion® Membranes and MEAs for PEM Water Electrolysers
    Stefano Giancola, Álvaro Reyes-Carmona, Marc Dupont, Sara Cavaliere, Eddy Moukheiber, Luca Merlo, Deborah Jones, Jacques Rozière
    Advances in Polymers for Fuel Cells and Energy Devices, Asilomar, February 2015

14. Advanced Electro-Catalysts and Membranes for proton exchange membrane water electrolysis.
    A.S. Aricò, S. Siracusano, V. Baglio, E. Moukheiber, L. Merlo

5th Regional Symposium on Electrochemistry South-East Europe, June 7-11 2015 Pravets, Bulgaria.

15. Challenges and Progress in Membranes, Catalysts, and MEAs for PEM Water Electrolysis
N. van Dijk, ECS Conference on Electrochemical Energy Conversion and Storage, Glasgow, July 2015

ELECTROHYPEM JOURNAL PUBLICATIONS

The Consortium has published, and will continue to submit for publication, a number of individual or joint publications to scientific journals. As for participation in international events, each publication has followed the dissemination protocol. Seven papers have been published in international journals during the lifetime of the project, and others including the most recent results will follow. The complete list is given in report D7.2, and on the project website.

1. *Electrochemical characterization of a PEM water electrolyzer based on a sulfonated polysulfone membrane*
S. Siracusano,V. Baglio, F. Lufrano, P. Staiti, A.S. Aricò,
J. Membr. Sci. (2013), 448, 209-214, DOI 10.1016/j.memsci.2013.07.058

2. *Design and testing of a compact PEM electrolyzer system*
N. Briguglio, G. Brunaccini, S. Siracusano, N. Randazzo, G. Dispenza, M. Ferraro, R. Ornelas, A.S. Aricò, V. Antonucci,
Int. J. Hydrogen Energy (2013), 38, 11519-11529, DOI 10.1016/j.ijhydene.2013.04.091

3. *Polymer electrolyte membrane water electrolysis: status of technologies and potential applications in combination with renewable power sources*
A. S. Aricò, S. Siracusano, N. Briguglio, V. Baglio, A. Di Blasi, V. Antonucci
J. Appl. Electrochem. (2013) 43, 107-118, *DOI 10.1007/s10800-012-0490-5*

4. *Proton exchange membrane water electrolysis with short-side-chain Aquivion membrane and IrO2 anode catalyst,*
A. Skulimowska, M. Zaton, M. Dupont, S. Sunde, L. Merlo, D. J. Jones, J. Rozère
Int. J. Hydrogen Energy, DOI 10.1016/j.ijhydene.2014.02.082

5. *Performance analysis of short-side-chain Aquivion perfluorosulfonic acid polymer for proton exchange membrane water electrolysis.*
S. Siracusano, V.Baglio, A.Stassi, L.Merlo, E.Moukheiber, A.S.Aricò.
J. Membr. Sci. 466(2014) 1–7, DOI 10.1016/j.memsci.2014.04.030

6. *Nanosized IrOx and IrRuOx electrocatalysts for the O2 evolution reaction in PEM water electrolysers.*
S. Siracusano, N. Van Dijk, E. Payne-Johnson, V. Baglio, A.S. Aricò.
Applied Catalysis B: Environmental 164 (2015) 488–495, DOI 10.1016/j.apcatb.2014.09.005

7. *Performance of a PEM water electrolyser combining an IrRu-oxide anode electrocatalyst and a short-side chain Aquivion membrane.*

S. Siracusano, V. Baglio, E. Moukheiber, L. Merlo, A.S. Aricò.
Int. J. Hydrogen Energy DOI/10.1016/j.ijhydene.2015.04.159

ELECTROHYPEM WORKSHOPS

Joint Workshop with SUSHGEN: A joint workshop was held with the Marie Curie ITN "Sustainable Hydrogen Generation" in November 2013. The programme included two invited presentations, presentations by the Marie Curie Fellows, and by several members of the Electrohypem partnership (listed below).

SUSHGEN Final Workshop - Joint Dissemination Event with Electrohypem, Paris, November 2013

1. Stack and system development for coupling PEM electrolysis with renewable power sources.
   Overview of the Electrohypem project,
   A. Di Blasi , N. Briguglio
2. Activities of ITM Power in Electrohypem,
   N. van Dijk
3. Aquivion membranes for fuel cells and electrolysers,
   E. Moukheiber

Dedicated Electrohypem Workshop: A PEM electrolysis workshop was organised (11th December 2014) as a side event of the Euro-Mediterranean Hydrogen Technologies Conference, Taormina

http://www.itae.cnr.it/emhytec2014/electrohypem.html

The focus of the ElectroHyPEM workshop inside the Emyhytec conference was to allow the stakeholders involved in hydrogen production by water electrolysis to present results on electrocatalysts, membranes, MEA, stack and systems development. Particular discussion was devoted to the integration of such systems with renewable energy sources in order to contribute to the road-map addressing the achievement of a wide scale decentralised hydrogen production infrastructure. The workshop participants included researchers, stakeholders, industry and institution representatives, providing oral or poster presentations and participating in the discussion on recent progress in the field.

Presentations made at the workshop by the partners are listed below:

ELECTROHYPEM Workshop - Taormina, December 2014

1. **Eddy Moukheiber** - Advanced fluorinated materials for low and high temperature PEM water electrolyser
2. **Nicholas Van Dijk** - Study of iridium ruthenium oxide structures for improved catalysis of the oxygen evolution reaction in a PEM electrolyser

3. **Antonino S. Aricò** - Enhanced performance and cost-effective materials for long-term operation of PEM water electrolysers coupled to renewable power sources
4. **Jose Manuel Subiñas Seco de Herrera** - Sustainable hydrogen production by PEM electrolysis for e-mobility
5. **Stefano Giancola** – Nanocomposite proton exchange membranes reinforced with electrospun polymer
6. **Stefania Siracusano** - Comparison of IrOx and IrRuOx oxides as anodic electrocatalysts for solid polymer electrolyte water electrolysis
7. **Georgios Tsotridis** - Discussion on harmonisation of testing protocols and procedures applied to electrolysis

FCH-JU Review Days

The coordinator has represented the project at each of the three FCH-JU review days during the duration of the project. Posters and/oral presentations were made as requested by the Programme Office.

ELECTROHYPEM Project oral presentation, Presenting Coordinator: Dr. Antonino Salvatore Aricò
FCH JU 2012 Programme review 28 & 29 November 2012, Brussels

ELECTROHYPEM Project poster, Presenting Coordinator: Dr. Antonino Salvatore Aricò
FCH JU Review Days 2013, 11 & 12 November 2013, Brussels

ELECTROHYPEM Project oral presentation, Presenting Coordinator: Dr. Antonino Salvatore Aricò
FCH JU Review Days 2014, 10 & 11 November 2014, Brussels

FUTURE DISSEMINATION PLANS

The consortium will continue to engage in conducting further activities for promoting and disseminating the project results. The following measures are planned so far in the near future to follow up the project:

-The website will be kept as a source of information source of the activities performed in the project. The website will also continue to receive and publish information on future published papers from the project.

Several publications and conferences are planned (D7.2, D7.3).

A.5. PROJECT WEB-SITE

http://www.electrohypem.eu/

Enhanced performance and cost-effective materials for long-term operation of PEM water electrolysers coupled to renewable power sources

ELECTROHYPEM is a project of the 7th European Framework Programme of the Fuel Cells and Hydrogen Joint Undertaking (FCH JU), running between 2012 and 2015.

The overall objective of the **ELECTROHYPEM** project is to develop cost-effective components for proton conducting membrane electrolysers with enhanced activity and stability in order to reduce stack and system costs and to improve efficiency, performance and durability.

The focus of the project is concerning mainly with low-cost electrocatalysts and membrane development. The project is addressing the validation of these materials in a PEM electrolyser for residential applications in the presence of renewable power sources.

The aim is to contribute to the road-map addressing the achievement of a wide scale decentralised hydrogen production infrastructure.

... Read more ....

The project website provides information on the project partnership, information on project progress meetings and dissemination activities. It provides links to the publishable abstract of published journal papers, and the full reference and DOI of each publication.

COORDINATOR CONTACT DETAILS

Dr. Antonino Salvatore Aricò
CNR-ITAE - Institute for Advanced Energy Technologies "Nicola Giordano"
Via Salita Santa Lucia Sopra Contesse, 5
I-98126 – Messina, Italy
Tel:+39-090624237
arico@itae.cnr.it