

# Premium Act (n°256776)

## First periodic report

### Publishable summary



# PREMIUM ACT

**Improvement of stationary PEFC systems durability (40000h required!)**  
 → A reliable method to predict system lifetime, benchmark components and improve operating strategies

**WP2 – Experiments under real operating conditions**

**WP3 – Quantification of components degradation**

**WP5 – Lifetime prediction methodology**

**WP1 - Specifications**

**WP4 – Predictive Modelling**

→ Validation of relevant **accelerated tests** able to couple various degradation factors and to assess different MEAs' lifetime more rapidly than with normal tests.

→ Final expected outcomes: **operating strategies** able to improve the lifetime of the DMFC & PEMFC systems studied and a **methodology** to predict the life time of their MEAs.

## **Publishable summary**

A general objective is to contribute to the improvement of stationary fuel cell systems durability, keeping in mind that the target required is 40000h. In this sense, the success of the project should help to overcome one of the main bottlenecks for European providers of stationary fuel cell systems and will contribute to cross cutting issues relevant for European R&D and fuel cell industry development. Thus, reliable systems corresponding to the technical specifications of the energy global market could be widespread, which will change the end-user habits towards the stationary energy management and will help to reduce greenhouse gas emission.

Premium Act specific objectives are to propose a reliable method to predict lifetime, based on validated accelerated degradation tests, to benchmark components and to improve operating strategies of real systems. The project addresses two types of technology: Direct Methanol and Proton Exchange Membrane Fuel Cells operating with reformat hydrogen.

As far as the degradation understanding is concern, focus of Premium Act is on the core of the fuel cells: the Membrane Electrode Assemblies (MEA). The structure of the project is based on the following scheme: initial information is requested from the real systems developed by the industry partners in order to define the reference conditions to which the MEA have to be submitted in controlled devices, namely short stacks and single cells, to check the nominal degradation of both the fuel cell performance and the components. Deep *in-situ* and *ex-situ* analyses are conducted to characterize the two latter types of degradation. In-situ tests are giving information about reversible or permanent performance degradation as well as electrochemical behaviour and properties modifications during ageing. Ex-situ methods are used to assess chemical, physical, structural properties of the components with a local resolution allowing to better relate degradation to heterogeneous conditions and operation in the stacks and cells. Segmented cell devices are also developed and used to directly relate local operating conditions to local performance. In order to enhance interpretation of non homogeneous operation and understanding of local degradation phenomena, modelling is used mainly to enable better description and prediction of coupling phenomena. Models are developed from electrochemical local level for mainly degradation mechanisms description to more macroscopic cell level with mainly fluid transport and current distribution simulation. The core idea of the project is to use the information coming from these degradation investigations to propose accelerate tests for the prediction methodology, and, operating strategies expected as the final results of the project.

For the first reporting period, the aim was mainly to start with these investigations of MEA performance and MEA components degradation in order to get as well experimental and modelling results as bases for reaching the final objectives during the second period.

Specific objectives for the period were related to specification, fuel cell testing, components characterization and modelling activities, always considering the two fuel cell technologies considered in the project: Direct Methanol and Proton Exchange Membrane.

During the first period, all reference components and conditions to be applied for ageing studies have been specified. Ageing tests have been conducted in several devices and conditions. Main results are that for both technologies, two types of degradation occur during ageing: reversible and permanent degradation. Work is conducted to reduce reversible degradation related to operating conditions, in order to be able afterwards to focus on permanent degradation for life time prediction methodology objective.

For DMFC, tests have been conducted in stationary and load cycling operation, impact of different parameters has been studied focusing on the major issue which is the methanol cross-over impacting on both electrodes efficiency. Most interesting result concerns the identification of refreshing conditions, allowing to reduce strongly overall reversible losses mainly due to degraded operation of the anode.

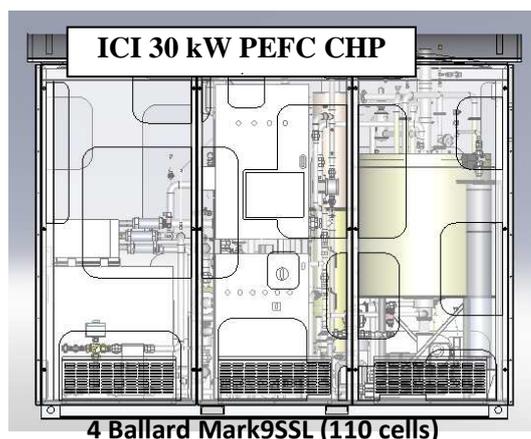
For PEMFC, main tests concern the impact of load cycles profile and of reformat fuel composition. Like for DMFC, the fuel cell shows mainly reversible degradation at least during 1000 hours of nominal cycles. It has been shown that carbon monoxide concentration in the reformat has a major influence on degradation rate and cell performance behaviour.

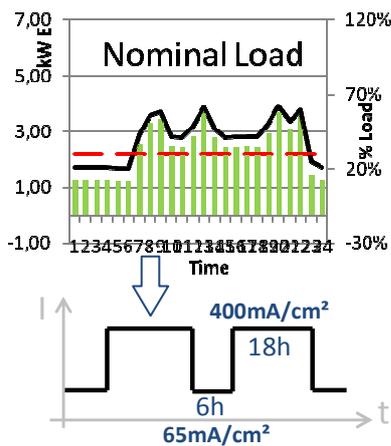
Many degradation analyses have been conducted by several spectroscopic techniques for chemical mapping of elements (in plane, through plane) and electron microscopy observation at different resolution and magnification. DMFC catalyst layers used here are very different of common Pt/C (or PtRu/C) catalysts, which is an interesting point because it could lead to potentially different degradation phenomena. Apart from catalyst particle growth often observed after fuel ageing, analyses of degraded MEA have allowed to identify that both technologies can lead to partial Ru dissolution (with evidence of some Ru cathode side for DMFC, and Ru element in the membrane for PEMFC). Analyses are on-going to check if mechanisms are impacted by local or global operating conditions. More accurate evidence of mechanisms are needed particularly to validate degradation models and allowing to integrate them in the performance description models.

Concerning modelling, the PEMFC cell model describing heterogeneous distribution of current density and fluidic parameters has been improved and coupled with degradation mechanisms coming from local electrochemical model. Simulation of performance evolution with this up-graded model has shown that ageing leads to an increase of initial heterogeneities of current distribution associated with heterogeneous active area distribution.

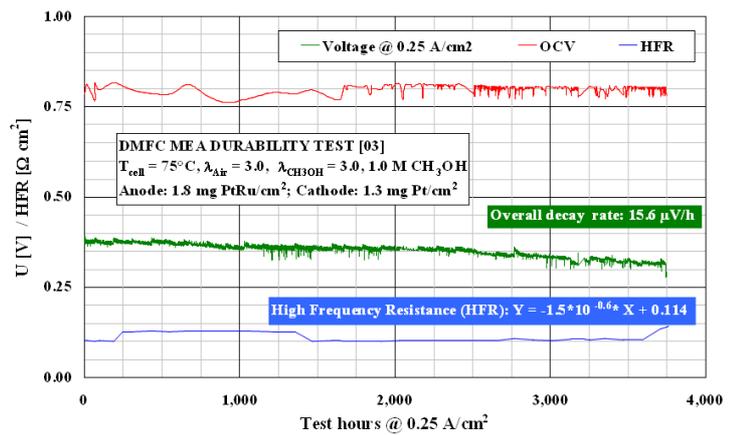
For DMFC modelling, a local model has been developed to describe Ru dissolution mechanism. Simulation has allowed to show preferential dissolution near the membrane. In parallel, a cell model has been improved with mainly water transport accurate description, because it is directly related to operation, homogeneity and hence degradation. The improved model has allowed to simulate water flooding. Next steps for modelling will be further combinations of degradation, fluidic, and local performance models for better interpretation of experimental degradation, including coupling effects and contribution to prediction methodology.

Core topic of the project has started to be addressed with first sensitivity studies on external parameters related to fuel composition, other operating conditions or load profiles. First insights about single or coupled features able to reduce performance temporary degradation or to increase and accelerate permanent degradation have been identified and will be deeply exploited in the second period of the project in order to complete final goals. Final expected outcomes are operating strategies able to improve the lifetime of the systems considered and a methodology to predict the life time of their MEAs. The latter will be based on relevant accelerated tests able to couple various degradation factors and to assess different MEAs' lifetime more rapidly than with normal tests. Core technical part of the second period will indeed be to combine all the information coming from the experimental tests or analyses and from the modelling to propose and validate these accelerated tests.

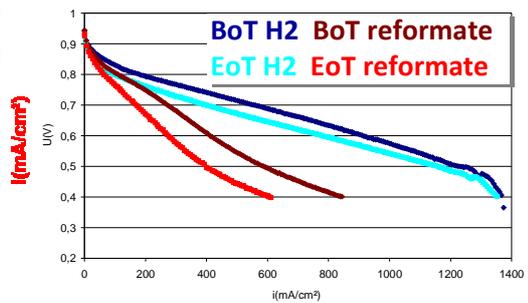
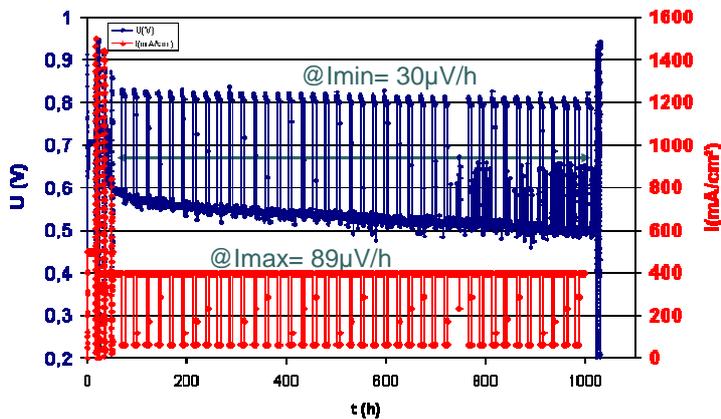




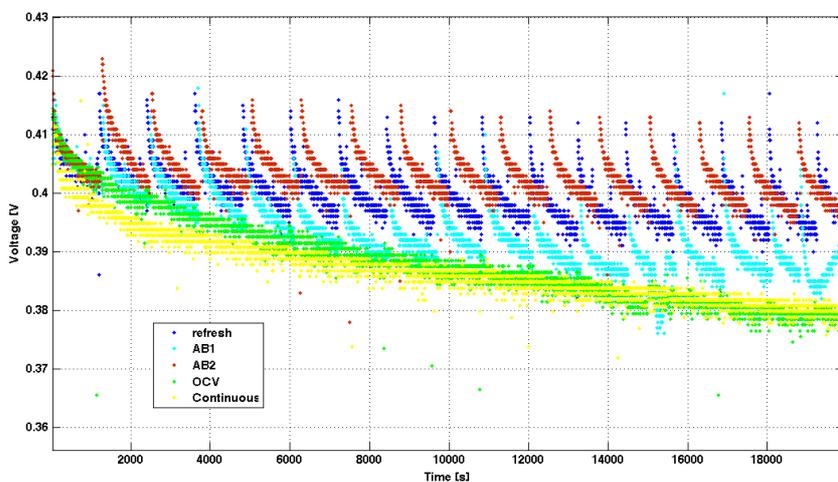
PEMFC load cycles selected by Soprano for stationary application



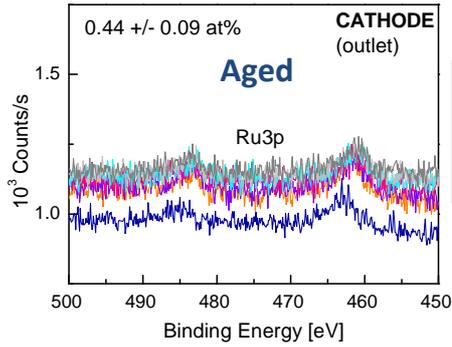
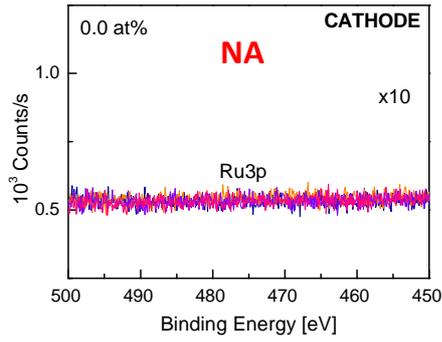
Long term operation of DMFC stack at IRD following real nominal condition



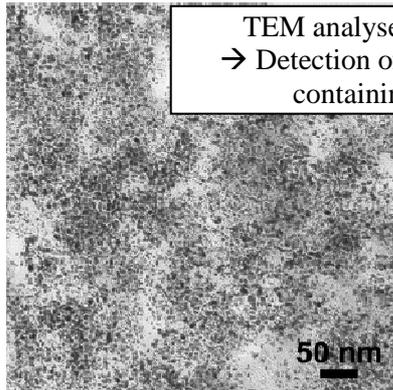
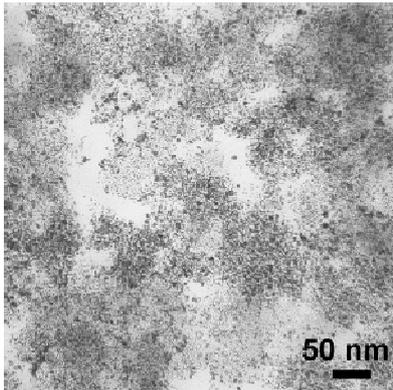
Long term test of PEMFC single cell at CEA following nominal load cycles profiles under critical fuel composition conditions condition



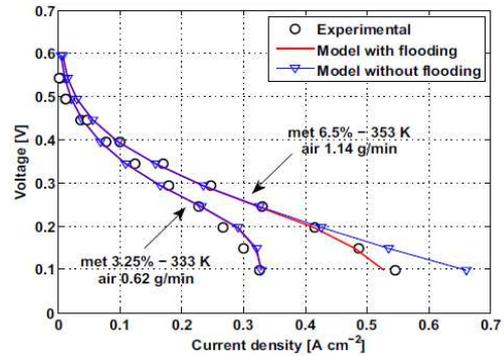
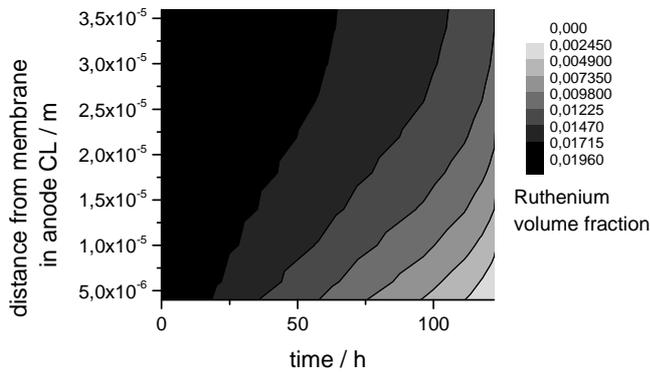
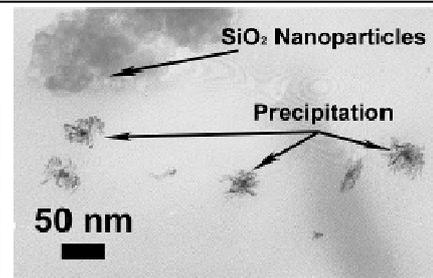
DMFC single cell operation at Polimi → Refresh cycles applied to reduce reversible performance degradation



XPS analyses of aged DMFC MEA at DLR  
→ Detection of Ru cathode side

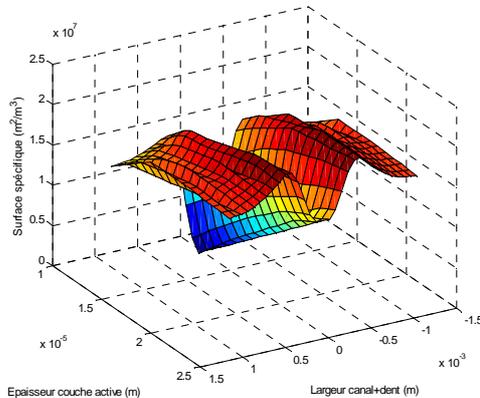
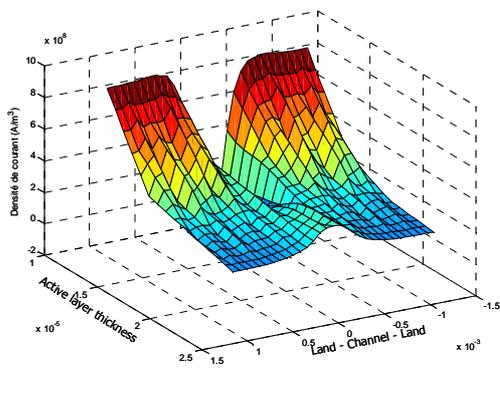


TEM analyses of aged PEMFC MEA at CEA  
→ Detection of Pt growth cathode side and of Ru containing particles in the membrane



DMFC local degradation model of DLR → Simulation of non homogeneous Ru dissolution

DMFC fluidic single cell model of Polimi → Simulation of flooding effect



PEMFC local performance model implemented with local degradation at CEA  
→ After ageing: increase of the current density heterogeneity and simulation of non homogeneous profile of Pt surface area