

Publishable summary

Proton Exchange Membrane Fuel Cells (PEMFCs) are complex nonlinear systems. In order to improve their durability and efficiency and to decrease their cost and required development time, the design of new diagnostic tools is crucial.

Powerful mathematical models of the dynamic behaviour of PEMFCs are necessary for such design and improvement of diagnostic tools. The project PUMA MIND will enhance the understanding of interactions, competitions and synergies among the mechanisms at multiple working scales from the material to the system level. It will lead to the development of robust dynamic macroscopic models for control-command purposes with predictive capabilities.

Most of the novel mathematical models developed by PUMA MIND have been tested experimentally, in order to ensure the applicability on commercial attainable components and catalysts. The most suitable catalysts for the MEA manufacturing technology will be used for these experiments.

The major target outcomes PUMA MIND are a set of simulation tools at the various scales between the material and the system level, which provide a better understanding of the interplay between mechanisms at different scales regarding the catalyst working (reaction mechanisms at the atomic scale), the electrochemistry (including degradation such as catalyst dissolution) and transport mechanisms (including water management), and their relative impact on the whole cell behaviour in real automotive application conditions (Figure 1).

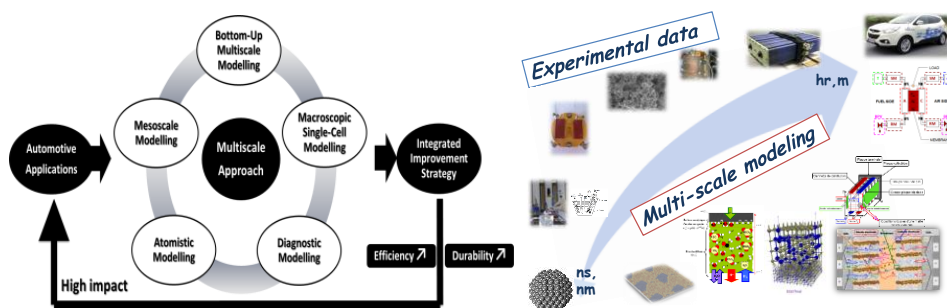


Figure 1: (a) Synergies within the PUMA MIND project to provide operation strategies to enhance the PEMFC performance and durability. (b) The multi-scale approach as well as experimental validation

To reach these challenging goals, the 11 partners from the PUMA MIND consortium are working in strong collaboration and are funded by the FCH-JU for 3 years (Figure 2).



Figure 2: The PUMA MIND consortium is co-funded by the FCH-JU

As showed in Figure 3, the project was organized in work packages (WPs), each technical WP is devoted to a given scale. Since the project aims to demonstrate a multiscale modeling methodology, strong interactions between the partners take place to ensure that the relevant inputs and outputs are achieved.

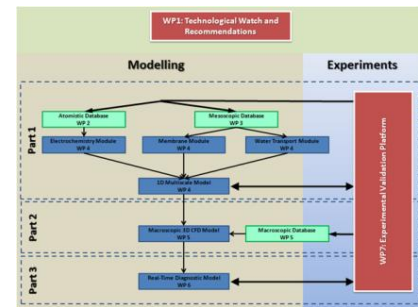


Figure 3: Pert diagram of PUMA MIND

Expected final results

The overall expected outcomes are:

- A detailed understanding of the multiscale interplaying and their impact on PEMFC performance and durability.
- The validation of the modeling codes with experiments by advanced characterization methods and in-situ diagnostic tools.
- The prediction of improved operating conditions, recovery protocols, material choice and component design.
- The demonstration of the feasibility of technical targets according to the DoE
 - Lifetime > 5000h
 - Efficiency > 55% at cell voltages > 700mV
 - Pt-loading < 0.15g/kW

Project public website

Public information about the project and its activities is made available via www.pumamind.eu

Project objectives, work progress and achievements

Project objectives for the second period

The objectives for the second period of PUMA MIND are in line with the description of work, and described below.

WP1: Specifications and Technology Watch

WP1 was almost completed during the first period of the project. Only Task 1.2 (Technology watch) continued in the second period. The aim of this task was to follow the advances in the field, which have been relevant for the project. In doing so, the consortium created a document in which all relevant papers which have been published since the start of the project are listed. For each of the papers a short description is provided in this document as well. In addition to this deliverable D1.2, the consortium decided to write a review paper based on the extensive literature research done within WP1 in order to share the gained knowledge with the community and also to increase the visibility of the PUMA MIND project. This comprehensive review paper has currently been published in the Journal of Power Sources.

WP2: Atomistic Modeling Platform

During the second period of the contract (18 months, 17/06/2014-16/12/2015), we have continued to investigate the adsorption properties of the ORR intermediates (mainly hydroxyl and hydroperoxyl species) and their associated reactivity on platinum nanoparticles from DFT calculations (Task 2.1). Activation Gibbs free energy barriers have been calculated in model environment conditions including temperature, solvation and bias voltage via two thermodynamic approaches that have been combined: the local solvation model and the computational hydrogen electrode model. The developed models and corresponding results and have thus gone beyond the initial scientific objectives. Several publications at the highest impact in the scientific community (Nature Chemistry & Science) have been obtained. In summary, the scientific objectives and associated deliverables of Task 2.1 have all been fulfilled. In a parallel way, we have developed other models related to the catalyst degradation (Task 2.2) and to the global effect of solvation on the adsorption properties (Task 2.3). Regarding the catalyst degradation, we have examined two different sintering models (extraction pathways of platinum atom from corner of nanoparticle or growth of platinum nanoparticle with adatoms) and a transport model (diffusion pathways of platinum atom over platinum nanoparticle facets). Kinetics of these processes (activation barriers and rate constants) have been calculated and the promoting effect of hydroxyl species has also been considered, as initially planned. The corresponding scientific objectives and deliverables of Task 2.2 have thus been all fulfilled. Concerning the solvent environment (Task 2.3), a global solvation model has been elaborated on the basis of an equilibrated interface between liquid water and platinum nanoparticle at 350 K through ab initio molecular dynamics. As initially planned, the adsorption of molecular oxygen and its dissociation have been studied. A comparison with the local solvation model has been carried out and all the scientific objectives of Task 2.3 have thus been fulfilled.

WP3: Mesoscale Level Modeling Platform

The main objective of this task is the development and implementation of models able to predict and describe the components and the interfaces structural properties from their chemical composition as well as their associated reactivity and transport properties. For that purpose, in LRCS proposes to develop a mesoscopic kinetic Monte-Carlo Code (KMC) devoted to study the catalyst reactivity and degradation process, based on the elementary kinetic reaction activation barriers obtained by DFT in WP2, in order to explore the ORR intermediates species coverage dynamic in the Inner Layer (IL). The outcomes of this model will be integrated in a multiscale simulation package called MS LIBER-T (*Multiscale Simulator of Lithium Ion Batteries and Electrochemical Reactor Technologies*), emerged in 2013, that integrates a new model of electrochemical double layers.

WP4: Full Bottom-Up Multiscale Simulations

The objectives of this during the second period concerned the integration of the data and mechanisms resolved in WP2 and WP3 into the pre-existing CEA full bottom-up multiscale model EDMOND. Intensive multiscale numerical simulations should then be performed in order to calculate the relative impact weight of the different mechanisms on the global cell performance and durability as function of the chemical and structural properties of the materials and components, and of the operation conditions. This will allow to better understand the interplaying of physicochemical and degradation mechanisms at the local scale and to help in the prediction of the performance and durability of the materials at the local level, and to provide guidelines about the processes to be incorporated in the models developed in WP5.

WP5: Macroscopic Single-Cell Models

The objectives of this workpackage during the second reporting period were to achieve the development of the 2D single cell CFD model to incorporate multicomponent performance and degradation functionality and to exploit the multiscale and multiplatform simulation platform to couple macroscopic single cell model and PUMA MIND degradation library. This allows to study the dependence of the global cell performance and durability on the chemical and structural properties of the materials and components and on the operation conditions. Based on these developments, the simulations will provide spatially averaged cell degradation data to WP6 for real-time diagnostics and control.

WP6: Real-Time Diagnostic Models

The goals of this workpackage during the second period was to continue developing and exploit the models and methods presented during the first reporting period. The first objective was to develop a control oriented model described by ordinary differential equations (ODE) based on the mathematical reduced version developed in WP6 for real-time diagnostic purposes. The second objective was to develop and implement on-board monitoring tools to determine fuel cell performance and degradation indicators based on the mathematical model. The final goal is the development of a model based control strategies with the purpose of enhancing the

PEMFC performance and durability, and to design dynamic observers for the estimation of states and performance variables in the PEMFC.

WP7: Experimental Validation Platform

The objective of the experimental validation platform during the second period concerns firstly *ex situ* experiments which will supply specific data to refine the description of the mechanisms in the electrochemical part of the WP4 model. These *ex situ* analyses will include specific electrochemical experiments such as RRDE and half-cells. Secondly, the SAS fuel cell and single-cell experiments will be performed for respectively WP4 and WP5 models validation for different operating conditions (reactant concentrations, temperature...) according to the protocols specified during the first reporting period. Finally, the electrochemical and structural TEM and XPS characterizations of the aged components will be achieved and data for the validation of the operation strategies based on the online control-command model developed should be provided.

WP8: Coordination of Scientific and Technical Strategy

No major problems of project management occurred in the second reporting period.

The grant agreement was signed by all the partners the 17th of December 2012.

WP9: Dissemination and exploitation

The consortium continued to promote the R&D results and impact of the project via a number of channels. These included presentations in over 55 scientific events and authoring of 62 publications (journal articles, book chapters and conference proceedings) as reported in D09.02. The project organised two workshops of its own. The first one in June 2014 at Minatec in Grenoble, France and the second one in March 2015 in Freiburg, Germany. Exploitation of results consists of 'leveraging' and 'mainstreaming' of results. The PUMA MIND exploitation plans contain partners' ideas about how to use the knowledge and tools acquired during the project at local, regional, national, European, and/or international levels. As the IP is deeply embedded in the fuel cell models, patents have not been considered as an attractive option. As per the consortium agreement where several partners have jointly carried out work generating foreground and where their respective share of the work cannot be ascertained, they shall have joint ownership of such foreground in equal parts. Given the level of maturity of the technology and integration of the different levels the decision taken is to focus on securing contract research, either from private or public sources (national, European, or international). In this case each of the joint owners shall be entitled to use the foreground as it sees fit, including but not limited to grant non-exclusive licenses to third parties, without any right to sub-license. The consortium believes that results should be of particular interest to automotive companies active in fuel cell vehicles as well as specialist OEMs. Importantly, few PUMA MIND researchers now work for such organisations facilitating technology transfer and supporting the formation of new partnerships.