Coupling experiments and modeling: towards a better understanding of PEMFC operation

Florent Vandenberghe, Pascal Schott, Fabrice Micoud, Arnaud Morin, Marian Chatenet

Coupling Experiments and Modeling: Towards a Better Understanding of PEMFC Operation

Florent Vandenberghe[1,*], Pascal Schott[1], Fabrice Micoud[1], Arnaud Morin[1], Marian Chatenet[2]
[1]*University Grenoble Alpes, CEA-LITEN, F-38054 Grenoble, France*
[2]*LEPMI, CNRS-University Grenoble Alpes-Grenoble INP, France*
**(florent.vandenberghe@cea.fr)*

Despite many improvements, PEMFC systems still suffer technological limitations, some of which being linked to the materials' cost, performance and durability of the cathode catalyst layer. Combining experiments and modeling enables to characterize and better understand the behaviour of the carbon-supported platinum (Pt/C) electrocatalyst, its utilization/effectiveness in PEMFC cathode catalyst layers and to be able to predict the performance and durability of PEMFC. In this work, physico-chemical and electrochemical measurements are performed from the scale of the raw Pt/C materials up to the complete catalyst layer, to gather as much information as possible on the catalytic layer micro-structure and its operating properties. During experimental performance measurements in PEMFC differential-single cell (1.8 cm^2 under high reactant stoichiometry), reversible performance gains/losses can be observed. This behaviour is often ascribed to variable hydration state of the ionomer in the catalyst layer and the bulk membrane and also to the evolution of platinum surface states due to oxides partial formation/reduction.

In parallel; several models have been developed to simulate one or combined mechanisms leading to the performance[1] and degradation of cell components[2,3]. Based on our experimental work and data sets, the behaviour of the Pt/C electrocatalysts has been studied in order to introduce new electrocatalytic features in one-dimensional models, especially the Pt surface oxide formation and reduction through basic reactions linked to the surface state of Pt as well as to 'bulk' Pt oxide formation via chemical place exchange reaction. This preliminary step was successfully implemented into a complete performance model for the O_2 reduction reaction at the cathode, that better describes the physical and electrochemical phenomena involved in very low loaded cathode catalyst layers during fuel cell operation. Hydration variation of the cathode catalyst layer and Pt oxide formation/reduction are now well described in the model during performance simulation leading to hysteresis phenomena as observed experimentally. Impact of the cathode catalyst layer structure, composition and loading as well as transport properties influence the hysteresis phenomenon and will be discussed.

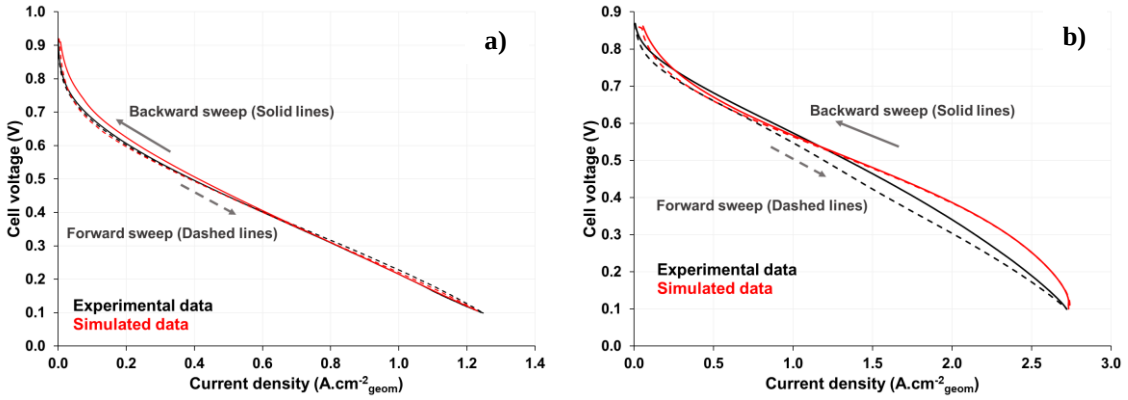

Figure1: Comparison between experimental and simulated performances measurement: ***a)*** *with 0.020 $mg_{Pt}.cm^{-2}$ loading at the cathode* ***b)*** *with 0.1 $mg_{Pt}.cm^{-2}$ loading at the cathode*

(1) Randrianarizafy, B. *et al.* Design Optimization of Rib/Channel Patterns in a PEMFC through Performance Heterogeneities Modelling. *Int. J. Hydrog. Energy* **2018**, *43* (18), 8907–8926. https://doi.org/10.1016/j.ijhydene.2018.03.036.
(2) Jahnke, T. *et al.* Physical Modeling of Catalyst Degradation in Low Temperature Fuel Cells: Platinum Oxidation, Dissolution, Particle Growth and Platinum Band Formation. *J. Electrochem. Soc.* **2019**, *167* (1), 013523. https://doi.org/10.1149/2.0232001JES.
(3) Maranzana, G. *et al.* Startup (and Shutdown) Model for Polymer Electrolyte Membrane Fuel Cells. *J. Electrochem. Soc.* **2015**, *162* (7), F694–F706. https://doi.org/10.1149/2.0451507jes.