

CATALYTIC LAYERS OF PROTON EXCHANGE MEMBRANE FUEL CELLS BASED ON CARBON XEROGEL SUPPORTS

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Carbon materials are commonly used as electrode components in electrochemical systems. Carbon black remains the most widespread support to prepare catalytic layers of Proton Exchange Membrane Fuel Cells (PEMFCs). The composition and the pore texture of these carbons are key variables to the device performance, through (i) the ability of the support to disperse metal atoms, (ii) the electrolyte/carbon contact and (iii) the reactant/product mass transport within the porosity [1]. However, in the case of carbon black, the mastering of these properties is not easy. In particular, the pore texture of the final electrode is not controllable since it largely depends on the electrode processing (*i.e.* electrode pressing), and not on the material itself.

Replacing these conventional carbons by nanostructured materials, such as carbon xerogels (CXs), allows for improving their performance and understanding the impact of the pore texture on the catalytic layer performance [2]. CXs are obtained by drying and pyrolysis of aqueous organic gels, and their pore texture can easily be tailored within a very wide range by adjusting the synthesis variables at an adequate value [3]. The final carbon is made of interconnected spherical-like microporous nodules, the size of which is controlled by the gel synthesis variables. As a result, one can obtain very pure carbons with meso-macropore sizes ranging from a few nm to a few μm , with pore volumes as high as $3 \text{ cm}^3/\text{g}$ and large specific surface area ($\sim 600 \text{ m}^2/\text{g}$). In addition, the micropore texture and surface chemistry can be modified via post-treatments in order to tune these properties independently [4, 5]. CXs are thus very interesting candidates to replace carbon blacks with a catalyst support with tunable (surface) chemical composition and adequate pore texture, which both impact the performance of PEMFC catalytic layers.

The synthesis of CX-supported Pt catalysts has been studied and optimized [6]. Pt catalysts with excellent dispersion (particle size $\sim 3\text{-}4 \text{ nm}$, Fig. 1a) and high mass fractions (min. 40% wt.) supported on texture-tailored carbon materials (pore size from 20 to 500 nm) were prepared using simple impregnation-reduction processes in liquid phase. Final catalysts are obtained from the powdery support and the metal precursor within about 1 h. They display excellent intrinsic catalytic activities and can be used to prepare Membrane-Electrode Assemblies (MEAs) with interesting mass transport properties compared to those elaborated from supported catalysts on carbon black.

The same type of nanostructured carbon can be used to synthesize supported hollow PtCo nanoparticles (Fig. 1b) [7]. The addition of Co leads to the Pt lattice contraction, and thus to much higher catalytic activity while the hollow structure allows for good metal utilization ratio despite the large particle size. These catalysts were thoroughly characterized using physico-chemical and electrochemical techniques, including Accelerated Stress Tests (ASTs) on rotating disk electrodes. The hollow PtCo/CX catalysts display much better stability than Pt/CX and Pt/carbon black samples, probably due to the particle size. PtCo/CX catalysts were also tested in Membrane-Electrodes Assembly configuration for at least 200 h, which allowed comparing the overall stability of the samples. ASTs were performed and results show that the stability of the hollow PtCo/CX catalysts are far more stable than Pt nanoparticles,

whatever the support used: indeed, while the performances of Pt nanoparticles continually decrease with ageing time, the current-voltage curves obtained with PtCo/CX hollow particles stabilize after a while.

Current research goes towards the modification of the carbon surface properties, for instance by nitrogen doping *via* plasma treatments. The goal is to improve the nanoparticle stability and increase further the catalysts lifetime.

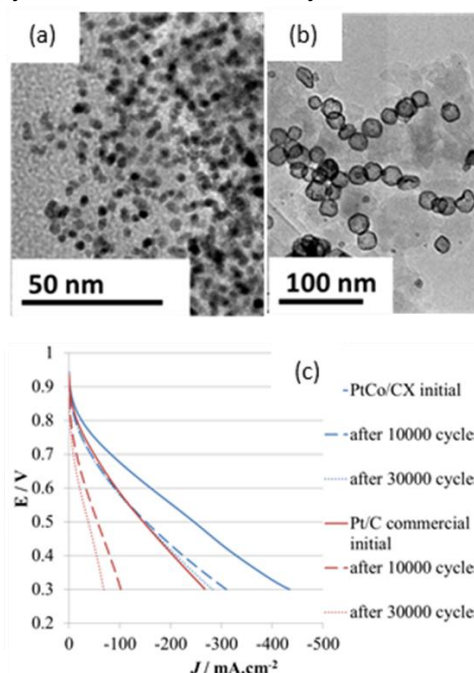


Figure 1. (a) TEM picture of a Pt/CX catalyst (40% wt.) prepared by impregnation-reduction process. (b) TEM picture of hollow PtCo/C nanoparticles. (c) Comparison between commercial Pt/carbon black and hollow PtCo/CX catalysts in MEA (H_2/O_2); the Pt content is constant ($0.033 \text{ mg}_{\text{Pt}}/\text{cm}^2_{\text{electrode}}$), and cells were cycled up to 30,000 times between 0.6 and 1.0 V to accelerate ageing.

Acknowledgements. NJ thanks the Walloon Region for funding (project HYLIFE, grant n° 1410135).

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