

Low-loading Pt, Hierarchical “Core-Shell” Electrocatalysts for the Oxygen Reduction Reaction Based on a Graphene “Core” and a Carbon Nitride “Shell”

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A major restructuring of the energy system is underway at the global level, with the purpose to curtail the dependence from fossil fuels and minimize the emissions of greenhouse gases [1]. A crucial role is to be played by electrochemical energy conversion and storage (EECS) systems, such as proton exchange membrane fuel cells (PEMFCs), that convert into electrical energy the chemical energy associated with the oxidation of hydrogen. One of the most important bottlenecks in PEMFC operation is the oxygen reduction reaction (ORR), that takes place at the PEMFC cathode and needs to be promoted by suitable electrocatalysts (ECs).

This work overviews the development of a new family of low-Pt ECs for the ORR. The active sites of the ECs are found on the surface of sub-nanometric clusters (SNCs) consisting of PtM_x alloys, where M is a first-row transition metal (e.g., Ni, Cu). M operates as a “co-catalyst” and raises the intrinsic performance of each active site much above the Pt baseline [2]

With respect to the Pt nanoparticles (NPs) adopted in state-of-the-art ORR ECs, the SNCs increase the utilization of Pt atoms included therein by up to ca. one order of

magnitude and make them much more available for electrocatalytic purposes.

The support of the low-Pt ECs described here exhibits a “core-shell” morphology; it comprises a hierarchical graphene-based (H-GR) “core” (including highly defected graphene nanoplatelets and carbon black NPs) that is covered by a carbon nitride (CN) “shell” [3].

The SNCs are stabilized on the EC surface by means of strong interactions with defects of the graphene nanoplatelets and C- and N-based ligands of the CN “shell”, making up the “coordination nests”. As a result, the low-Pt ECs described here exhibit an outstanding durability.

This work takes into consideration the various families of low-Pt ECs based on H-GR supports developed so far by our group, and discusses extensively the interplay between preparation parameters, physico-chemical properties and electrochemical behavior, comparing results with state-of-the-art ECs.

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References

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