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Chemically functionalized carbon nanotubes with pyridine groups as easily tunable N-decorated nanomaterials for the oxygen reduction reaction in alkaline medium

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Abstract

We report on the N-decoration of multiwalled carbon nanotubes (MWCNTs) via chemical functionalization under mild reaction conditions. The introduction of tailored pyridinic functionalities as N-containing edge-type group mimics generates effective catalysts for the oxygen reduction reaction (ORR) in an alkaline environment. The adopted methodology lists a number of remarkable technical advantages, among which is an easy tuning of the electronic properties of N-containing groups. The latter aspect further increases the level of complexity for the rationalization of the role of the N-functionalities on the ultimate electrochemical performance of the as-prepared metal-free catalysts. Electrochemical outcomes crossed with the computed electronic charge density distributions on each scrutinized pyridine group have evidenced the central role played by the N-chemical environment on the final catalyst performance. Notably, small variations of the atomic charges on the N-proximal carbon atoms of the chemically grafted heterocycles change the overpotential values at which the oxygen reduction reaction starts. The protocol described hereafter offers an excellent basis for the development of more active metal-free electrocatalysts for the ORR. Finally, the as-prepared catalytically active materials represent a unique model for the in-depth understanding of the underlying ORR mechanism. © 2014 American Chemical Society.

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