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Chemical functionalization of N-doped carbon nanotubes: A powerful approach to cast light on the electrochemical role of specific N-functionalities in the oxygen reduction reaction

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Abstract

© 2016 The Royal Society of Chemistry. In this paper, we describe the combination of two different synthetic approaches to carbon nanotube N-decoration/doping: the chemical functionalization with tailored N-pyridinic groups and the classical Chemical Vapor Deposition (CVD) technique. Accordingly, CVD-prepared N-doped CNMs (NMWs) and their N-decorated (chemically functionalized) counterparts (NMW@N_{1,2}) have been prepared and used as metal-free electrocatalysts for the oxygen reduction reaction (ORR). It has been demonstrated that chemical functionalization occurs on the NMW surface sites responsible for their inherent electrochemical properties and "switches them off". As a result, the ORR promoted by NMW@N_{1,2} is fully controlled by the appended N-heterocycles. A comparative analysis of N-functionalized samples and N-doped (CVD prepared) materials is used to foster the hypothesis of a unique N-configuration (N-pyridinic) responsible for the overall electrochemical performance in NMWs. In addition to that, original electrochemical insights unveiled during the study are discussed and the truly metal-free action of NMW in ORR catalysis is demonstrated.

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