Revealing the Origin of Activity in Nitrogen-Doped Nanocarbons towards Electrocatalytic Reduction of Carbon Dioxide - DTU Orbit (09/11/2017)

Revealing the Origin of Activity in Nitrogen-Doped Nanocarbons towards Electrocatalytic Reduction of Carbon Dioxide

Carbon nanotubes (CNTs) are functionalized with nitrogen atoms for reduction of carbon dioxide (CO_2). The investigation explores the origin of the catalyst's activity and the role of nitrogen chemical states therein. The catalysts show excellent performances, with about 90% current efficiency for CO formation and stability over 60 hours. The Tafel analyses and density functional theory calculations suggest that the reduction of CO_2 proceeds through an initial rate-determining transfer of one electron to CO_2, which leads to the formation of carbon dioxide radical anion (CO_2C). The initial reduction barrier is too high on pristine CNTs, resulting in a very high overpotentials at which the hydrogen evolution reaction dominates over CO_2 reduction.The doped nitrogen atoms stabilize the radical anion,thereby lowering the initial reduction barrier and improving the intrinsic activity. The most efficient nitrogen chemical state for this reaction is quaternary nitrogen, followed by pyridinic and pyrrolic nitrogen.

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