Abstract

Carbon-based nanomaterials, particularly N-doped graphene derivatives, offer a viable and economical alternative to noble metal catalysts for the oxygen reduction reaction (ORR) in proton exchange membrane fuel cells (PEMFCs). This research addresses different synthesis methods and provides in-depth morphological, chemical and electrochemical analysis of various heat-treated graphene and N-doped graphene oxide derivatives, focusing on N-doped graphene oxide (N-htGO) and N-doped graphene oxide nanoribbons (N-htGONR). Work focuses on the investigation, validation, and comparison of factors influencing the ORR performance. Such as the pH dependent effect of inherent metal impurities, the aspect ratio contrast between 2D graphene and quasi-1D nanoribbons dimensions, considerations, improved surface area, and the influence of N-configuration on ORR activity. Based on results, the most promising properties are comprehensively integrated into N-doped graphene derivatives (N-GOD) and evaluated in a PEMFC for ORR.

Pushing the boundaries of carbon-based materials research, we introduce an innovative, scalable method of electrical induction heat treatment. This innovative approach not only accelerates the reaction process, but also enhances the energy efficiency and ORR performance of N-GOD in acidic and alkaline media.

At the same time, we investigated the potential of N-GOD and undoped GOD for green electrochemical conversion of ORR to hydrogen peroxide (H_2O_2), a sustainable alternative to the traditional anthraquinone autooxidation method. In particular, Ni-based heat-treated graphene oxide (Ni@htGO) materials showed remarkable ORR selectivity. To additionally improve the efficacy of Ni@htGO, we investigated various temperature-controlled synthesis routes with different nickel configurations, revealing a link between Ni functions and H_2O_2 production.

In summary, this comprehensive study sheds light on the synthesis, characterization, optimization, and efficacy of non-metallic N-htGOD electrocatalysts, either for H_2O_2 production or PEMFCs applications.

Key words: N-doped graphene derivatives, oxygen reduction reaction, proton-exchange membrane fuel cells, hydrogen peroxide production, metal impurities