

Vol. 33 • No. 18 • March 25 • 2026

www.small-journal.com

NANO · MICRO small



WILEY-VCH

RESEARCH ARTICLE

Heteroatom Loading of Nanodiamonds Modulates the Coupled Electrocatalytic Production of H₂O₂ by Oxygen Reduction and Water Oxidation

Xiaojie Jia | Jinjie Zhang | Yihua Zhu | Haibo Jiang | Jianhua Shen

Shanghai Engineering Research Center of Hierarchical Nanomaterials, Key Laboratory For Ultrafine Materials of Ministry of Education, Frontiers Science Center for Materialobiology and Dynamic Chemistry, School of Materials Science and Engineering, East China University of Science and Technology, Shanghai, China

Correspondence: Haibo Jiang (jianghaibo@ecust.edu.cn) | Jianhua Shen (jianhuashen@ecust.edu.cn)

Received: 10 October 2025 | Revised: 16 January 2026 | Accepted: 20 January 2026

Keywords: electrocatalytic H₂O₂ synthesis | heteroatom loading | nanodiamonds | two-electron oxygen reduction | two-electron water oxidation

ABSTRACT

The synthesis of H₂O₂ via two-electron oxygen reduction (2e[−] ORR) and two-electron water oxidation (2e[−] WOR) has attracted attention due to its simplicity, energy efficiency, and sustainability. However, the electrochemical conversion of H₂O to H₂O₂ often suffers from limited selectivity at high current densities. Herein, we propose oxygen-doped nanodiamonds (O-ND) for 2e[−] ORR to investigate the combined effects of heteroatom doping and the advantageous *sp*³ conformation on catalytic activity. We comprehensively characterized the configuration, doping, and crystal growth of O-ND using various techniques. The oxygen-doped *sp*³ diamond structure exhibited high catalytic activity for 2e[−] ORR, with Faradaic efficiency (FE) increasing from 71% in pristine ND to 87.1% and an average FE of 80%. We also synthesized copper-doped nanodiamond composite catalysts (Cu-OND) with high 2e[−] WOR activity, achieving 2e[−] WOR FE of 78.2%. The complete battery system, after coupling O-ND (cathode) with Cu-OND (anode), demonstrates outstanding performance at 1.8 V, with the combined FE of cathode and anode reaching 161.1%. This result surpasses most currently reported values. Density functional theory (DFT) calculations revealed that the maximum dynamic active sites of the coupled reaction catalyst are derived from the local charge regulation effect of O-doped *sp*³C and the heterojunction structure of Cu-doped OND.

1 | Introduction

Since its initial discovery and synthesis over two centuries ago, hydrogen peroxide (H₂O₂) has emerged as a vital chemical compound with extensive applications across diverse domains, including the chemical industry, environmental protection, and medical sciences [1]. The traditional industrial production method for H₂O₂ is the anthraquinone process [2]. Although this is a technically mature H₂O₂ production technology, it requires high energy input and generates organic waste during the reaction process, which is detrimental to environmental protection [3]. Two-electron oxygen reduction (2e[−] ORR;

O₂ + 2H⁺ + 2e[−] → H₂O₂, E₀ = 0.69 V vs. RHE) and two-electron water oxidation (2e[−] WOR; 2H₂O → H₂O₂ + 2H⁺ + 2e[−], E₀ = 1.76 V vs. RHE) are two pathways for H₂O₂ production. Due to their simplicity, low energy consumption, and alignment with sustainable development principles, they have become research hotspots.

Recent pioneering studies have reported various high-performance catalysts for 2e[−] ORR, including noble metal-based materials [4–8], transition metal-based materials [9–12], carbon-based nanomaterials [13–18], and others [19]. In contrast to 2e[−] ORR, 2e[−] WOR does not depend on gaseous reactants and solely