

International conference

Functional Inorganic Materials



FIM 2022

Abstract book



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<https://doi.org/10.15388/Proceedings.2022.29>

ISBN 978-609-07-0777-7 (digital PDF)



International Conference
Functional Inorganic Materials 2022

Sponsors



Can Graphene be Sensitive?

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Graphene and its oxide (GO) due to its properties are widely applied in electrochemical research as an electrode material. The immobilization of GO and its related nanomaterials into electrochemical sensors has shown great promise due to its high surface area, chemical stability, electron transfer rate, and easy functionalization. However, the field of graphene-based sensors is still in its early development stage, and several key challenges need to be addressed. The current progress has proved that the doping of heteroatoms (such as N, P, and B) in the graphene lattice is a feasible method to modulate the surface chemistry and electronic properties of graphene [1]. Therefore, this work aimed to produce N-doped reduced graphene oxide (rGO) and investigate its suitability in electrochemical sensing.

In this work, GO was prepared from natural graphite using the synthesis protocol reported by Yan et al. [2]. The obtained pre-oxidized graphite was subjected to oxidation by Hummers' method using NaNO₃, H₂SO₄, and KMnO₄ [3]. The rGO was produced from GO using a thermal shock method. The dried GO powder was quickly inserted into a preheated tubular furnace at a temperature of about 800 °C in the Ar atmosphere. To introduce N-functionalities, the rGO surface was modified with gaseous ammonia at temperatures of 850 °C and 950 °C for 4 and 8 h, respectively [4]. To achieve a complete comprehension of the effect on N-doped rGO structure, the nature of N-functionalities introduced during the functionalization and composition, a combination of various analysis methods such as Raman spectroscopy, X-ray photoelectron spectroscopy, SEM investigations, were used. Electrochemical measurements, in particular, cyclic voltammetry and chronoamperometry were used to evaluate the sensitivity of the obtained samples toward H₂O₂ and glucose detection.

The results demonstrated that the amount and type of N-containing functional groups introduced during the functionalization of rGO surface plays a crucial role in controlling the structure and the application potential of rGO. Moreover, it was observed, that various nitrogen species including pyridinic-N, pyrrolic-N, and quaternary-N were detected in the N-doped rGO. Also, it was demonstrated, that the N-doping at higher temperatures and longer time dramatically interrupts the carbon lattice and causes a high defective degree. Electrochemical studies showed that the attachment of a greater content of quaternary nitrogen species onto the rGO surface significantly improves electrocatalytic activity toward H₂O₂ reduction and glucose oxidation. The analytical performances of such sensors will be presented and discussed.

Acknowledgments

This project has received funding from European Social Fund (project No 09.3.3-LMT-K-712-19-0050) under grant agreement with the Research Council of Lithuania (LMTLT).

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