66TH INTERNATIONAL

OPEN READINGS

CONFERENCE FOR STUDENTS OF PHYSICS AND NATURAL SCIENCES

ANNUAL ABSTRACT BOOK | 2023

Vilnius University

VILNIUS UNIVERSITY PRESS

Editors

Martynas Keršys
Šarūnas Mickus

Cover and Interior design
Milda Stancikaitė

Vilnius University Press
9 Saulėtekio Av., III Building, LT-10222 Vilnius
info@leidykla.vu.lt, www.leidykla.vu.lt/en/
www.knygynas.vu.lt, www.journals.vu.lt

Bibliographic information is available
on the Lithuanian Integral Library Information System (LIBIS) portal ibiblioteka.lt.

ISBN 978-609-07-0883-5 (ePDF)

DOI: https://doi.org/10.15388/IOR2023

NITROGEN-MODIFIED REDUCED GRAPHENE OXIDE AS A PLATFORM FOR SENSITIVE AND SELECTIVE ELECTROCHEMICAL DETECTION OF DOPAMINE

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Dopamine (DA) is one of the most important neurotransmitters in the human body, playing a crucial role in motor control, reward and motivation functions, as well as the central nervous and immune systems. Abnormal levels of DA may potentially lead to several neurological diseases such as schizophrenia, depression, attention deficit hyperactivity disorder (ADHD), Parkinson's and Alzheimer's diseases [1]. Therefore, there is an urgent need to develop new platforms for the precise and sensitive determination of DA concentrations in the human body.

In recent years, electrochemical methods have been considered as an efficient tool for DA detection because of their simplicity, cost-effectiveness, fast response time, and reproducibility. Graphene-based derivatives such as graphene oxide (GO) and reduced graphene oxide (rGO) have drawn increasing attention as electrode materials in the development of electrochemical sensors due to the time-efficient and non-expensive synthesis procedure as well as unique chemical, physical, and electronic characteristics. rGO-based materials exhibit a high surface area, chemical stability, electrical conductivity, and the ability to immobilize a variety of different molecules [2]. However, despite these advantages, the development of rGO-based sensors with high sensitivity and rapid response time is still challenging. Therefore, doping with heteroatoms could be a key strategy to tailor structural and electrochemical properties of rGO because the doping process can enhance the charge carrier density, wettability, sensitivity, create new electrocatalytically active sites, and ensure a fast electrochemical response for accurate and sensitive DA detection [2,3].

The aim of this study was to synthesize N-doped reduced graphene oxide (N-rGO) samples and investigate their electrochemical performance in the detection of DA. GO has been prepared using a modified Hummers' method, including the pre-oxidation of natural graphite powder by the mixture of $K_2S_2O_8/P_2O_5/H_2SO_4$ [4]. N-rGO samples have been synthesized by a facile and one-pot hydrothermal treatment of GO in the presence of the organic dye "Bismarck Brown Y" (BB), which has been used as a N source for the rGO modification for the first time. Obtained samples (rGO, rGO_BB20, and rGO_BB50) have been characterized by Brunauer-Emmett-Teller (BET) analysis, scanning electron microscopy (SEM), X-ray photoelectron (XPS), and Raman scattering spectroscopies. The electrical behavior of the prepared materials has also been determined by estimating the dependence between electrical conductivity and bulk density. Electrochemical measurements, including cyclic voltammetry (CV) and chronoamperometry (CA), have been performed to analyse the sensitivity of N-rGO samples toward DA detection. Interference studies have also been carried out to investigate the selectivity of N-rGO samples.

The results of the XPS analysis show that rGO_BB20 and rGO_BB50 consist of 5.3 and 14.2 at% of nitrogen, respectively, indicating a successful rGO modification with N atoms. From the N1s XPS spectra it has been observed that in the rGO_BB20 sample nitrogen occurs mostly in the form of the pyridinic groups (45.0 at%), whereas in the rGO_BB50 sample the pyridinic (40.7 at%) as well as amine groups (35.7 at%) are dominant. I_D/I_G values determined from Raman spectra confirm the increase of structural disorders after the use of the BB additive. The BET analysis shows that the specific surface area decreases with the higher amount of BB used in the hydrothermal synthesis. Electrical conductivity measurements demonstrate that rGO and rGO_BB20 exhibit similar electrical conductivity values, while rGO_BB50 has much lower. From the SEM images, it is found that modification with N atoms leads to corrugation of rGO layers. The CV studies reveal that the rGO_BB20 and rGO_BB50 samples exhibit prospective electrocatalytic activity toward the DA redox peak. The CA studies show that the proposed sensor based on rGO_BB20 demonstrates a relatively high sensitivity of 0.46 $\mu A\ \mu M^{-1}\ cm^{-2}$ and a low limit of detection (11 nM). Interference analysis in the presence of other electroactive materials such as uric acid, ascorbic acid, citric acid, and H_2O_2 shows the electrochemical response only for dopamine using the rGO_BB20-based electrode, suggesting that it is a promising electrode material for the sensitive and selective determination of DA.

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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