Newcastle University ePrints

Popescu S, Dale C, Keegan N, Ghosh B, Kaner R, Hedley J.

Rapid Prototyping of a Low-cost Graphene-based Impedimetric Biosensor.

Procedia Technology 2017, 27, 274-276.

Copyright:

© 2017 The Authors. Published by Elsevier Ltd. This is an open access article under the CC BY-NC-ND license http://creativecommons.org/licenses/by-nc-nd/4.0/ Peer-review under responsibility of the organizing committee of Biosensors 2016

DOI link to article:

https://doi.org/10.1016/j.protcy.2017.04.116

Date deposited:

21/07/2017

This work is licensed under a

Creative Commons Attribution-NonCommercial-NoDerivatives 4.0 International licence

Newcastle University ePrints - eprint.ncl.ac.uk

Available online at www.sciencedirect.com

ScienceDirect

Procedia Technology 27 (2017) 274 – 276

Procedia Technology

Biosensors 2016

Rapid prototyping of a low-cost graphene-based impedimetric biosensor

Sinziana Popescu[a], Carl Dale[b], Neil Keegan[b], Biswajit Ghosh[c], Richard Kaner[d], John Hedley[a]

[a]*School of Mechanical and Systems Engineering, Newcastle University, Newcastle-upon-Tyne, NE1 7RU, United Kingdom*
[b]*Institute of Cellular Medicine, Newcastle University, Newcastle-upon-Tyne, NE2 4HH, United Kingdom*
[c]*School of Energy Studies, Jadavpur University, Kolkata 700 032, India*
[d]*Department of Chemistry and Biochemistry, Los Angeles, CA 90095, United States*

Abstract

This paper presents a preliminary investigation towards rapid prototyping of a low-cost biosensor based on reduced graphene oxide (rGO). The devices are fabricated via a laser scribing process and their functionality is demonstrated by their functionalization and subsequent immobilization of 7% bovine serum albumin (BSA). Non-faradaic electrochemical impedance spectroscopy (EIS) indicated a 33-42% decrease in impedance upon immobilization. An electroless nickel deposition process is demonstrated to enable electrical contacts to the device, with optimized plating conditions (pH, temperature) leading to a rGO-nickel contact resistance of 19 Ω/mm^2.

© 2017 The Authors. Published by Elsevier Ltd. This is an open access article under the CC BY-NC-ND license (http://creativecommons.org/licenses/by-nc-nd/4.0/).

Peer-review under responsibility of the organizing committee of Biosensors 2016

Keywords: graphene; reduced graphene oxide; impedimetric biosensor; interdigitated electrode array; electroless nickel deposition

1. Introduction

Since its discovery in 2004 [1], graphene has received significant research attention due to its extraordinary properties [2]. Reduced graphene oxide (rGO) has been shown to be suitable for bio/sensing applications [3,4] due to its chemically active nature based on the presence of surface functional groups. However, large-scale production, effective electrical contacting and specific surface functionalization remain key issues for graphene development towards commercial biosensing applications. This work investigates a novel approach to low-cost rapid prototyping of functionalised graphene interdigitated electrode arrays (IDE) utilizing laser reduced graphene oxide on flexible substrates. To complete the sensor fabrication, a novel approach for low contact resistance interfacing via electroless nickel metallization is proposed.

2212-0173 © 2017 The Authors. Published by Elsevier Ltd. This is an open access article under the CC BY-NC-ND license (http://creativecommons.org/licenses/by-nc-nd/4.0/).
Peer-review under responsibility of the organizing committee of Biosensors 2016
doi:10.1016/j.protcy.2017.04.116

2. Results

The rGO sensing platform, containing 10 parallel rGO electrodes (6 mm long, 0.5 mm wide), was produced by employing a Lightscribe DVD drive (Figure 1A), as reported by El-Kady et al [5]. The material was characterized using XPS and Raman spectroscopy, confirming the efficient reduction of the graphene oxide. The rGO atomic composition consisted of 96% carbon and 4% oxygen species, with a reduction factor of ≈14, whilst the Auger peak D-parameter of 20.2 is indicative of the dominant sp^2 domain [6,7]. High resolution helium ion microscopy (HIM) validated the lateral exfoliation of the graphene layers upon the photo-thermal reduction process (Figure 1C). The average rGO sheet was 300 Ω/□, measured using a Hall probe station.

The rGO sensing platform was covalently functionalized using carboxyl-reactive EDC-NHS chemistry, which was confirmed using a quantum dot labeling technique. A good distribution of the deposited amino-coated CdSe quantum dots was observed, especially towards the track edges where oxygen species were present. Upon the successful functionalization with EDC-NHS, 7% bovine serum albumin (BSA) was immobilized on the rGO sensing platform. Non-faradaic impedance measurements (phosphate buffered saline PBS, pH 7.5) were taken between successive chemical modification steps. The impedance decreased as a result of the immobilization process (Figure 1B), modelled by the addition of a series capacitive element (in the equivalent circuit). Also, a significant decrease in the equivalent constant phase element was noted whilst the rGO porosity led to an inductive behaviour at high frequencies.

To complete the sensor platform, a two-step electroless nickel deposition was performed on the rGO surface, involving activation (palladium chloride) and deposition in a Ni:P metal bath (nickel sulfate). Optimized bath conditions were determined to be 65°C and pH of 7.3 for a 2 minute deposition time. The deposited film selectively covered the IDE rGO pads (without the inter-pad GO) and it contained 85% nickel (Figure 1C), quantified with EDX. The contact resistance of nickel onto the rGO was measured using the transmission line method as 19 Ω/mm^2.

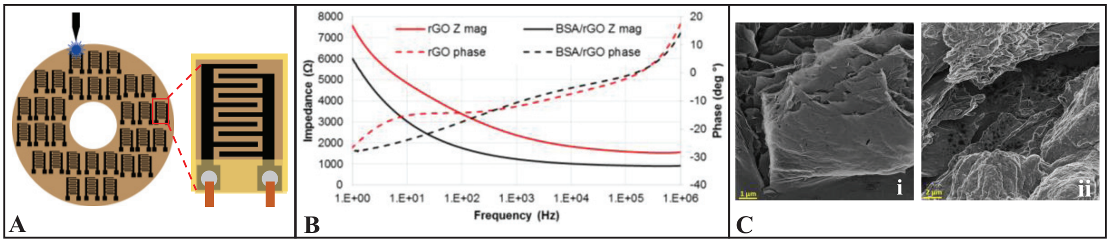

Figure 1: A) Lightscribe rGO production process and IDE diagram; B) Bode plot (impedance magnitude and phase) for the rGO biosensing platform before and after BSA immobilization; C) HIM image of (i) rGO and (ii) rGO after electroless nickel metallization.

3. Conclusions

Using the Lightscribe photo-thermal reduction method, rGO IDEs were efficiently manufactured on flexible substrates. The produced rGO showed good electro-chemical properties which are applicable towards biosensing applications and electroless nickel metallization techniques, presenting advantages towards low-cost, low temperature and scalable production of the sensor. Further work will address full biosensor integration and performance characterization.

Acknowledgements

The authors would like to thank EPRSC for financial support for the work (EP/I015930/1), the Leverhulme Trust (VPI-2013-003) and the British Council (UKIERI-TRP-2012/13-0010) for supporting this collaborative research between institutions.

References

[1] K. S. Novoselov, A. K. Geim, S. V Morozov, D. Jiang, Y. Zhang, S. V Dubonos, I. V Grigorieva, and A. A. Firsov, “Electric field effect in atomically thin carbon films.,” *Science*, vol. 306, no. 5696, pp. 666–669, 2004.

[2] X. Huang, Z. Yin, S. Wu, X. Qi, Q. He, Q. Zhang, Q. Yan, F. Boey, and H. Zhang, “Graphene-based materials: Synthesis, characterization, properties, and applications,” *Small*, vol. 7, no. 14, pp. 1876–1902, 2011.

[3] Y. Shao, J. Wang, H. Wu, J. Liu, I. A. Aksay, and Y. Lin, “Graphene Based Electrochemical Sensors and Biosensors: A Review,” *Electroanalysis*, vol. 22, no. 10, pp. 1027–1036, 2010.

[4] K. Griffiths, C. Dale, J. Hedley, M. D. Kowal, R. B. Kaner, and N. Keegan, “Laser-scribed graphene presents an opportunity to print a new generation of disposable electrochemical sensors,” *Nanoscale*, vol. 6, no. 22, pp. 13613–13622, 2014.

[5] M. F. El-Kady, V. Strong, S. Dubin, and R. B. Kaner, “Laser Scribing of High-Performance and Flexible Graphene-Based Electrochemical Capacitors,” *Science*, vol. 335. pp. 1326–1330, 2012.

[6] A. J. Barlow, O. Scott, N. Sano, and P. J. Cumpson, “Multivariate Auger Feature Imaging (MAFI) - a new approach towards chemical state identification of novel carbons in XPS imaging,” *Surf. Interface Anal.*, vol. 47, pp. 173–175, 2015.

[7] A. J. Barlow, S. Popescu, K. Artyushkova, O. Scott, N. Sano, J. Hedley, and P. J. Cumpson, “Chemically specific identification of carbon in XPS imaging using Multivariate Auger Feature Imaging (MAFI)”, doi: 10.1016/j.carbon.2016.05.073, Carbon N. *Y.*, 2016.