O14 |

UV PHOTODETECTOR BASED ON REDUCED GRAPHENE OXIDE AND n-TYPE Si HETEROJUNCTION

J.L. Pérez Gordillo[1], A. Meléndez[1], J.J. Santiago-Aviles[2], N.J. Pinto[1], I. Ramos[1]
[1]*Department of Physics and Electronics, University of Puerto Rico at Humacao, Humacao, Puerto Rico, 00791*
[2]*Department of Electrical and Systems Engineering, University of Pennsylvania, Philadelphia, PA 19104*
[*]*e-mail: idalia.ramos@upr.edu*

Introduction

The electrical, mechanical, thermal, and optical properties of reduced graphene oxide (rGO) make it an attractive material for the fabrication of photodetectors and other optoelectronic devices. In comparison with graphene, rGO can be prepared using inexpensive materials and simpler fabrication methods with high yields [1]. The properties of this material can be tuned with the precursor solution, synthesis, and reduction processes. A key to the production of high quality rGO is the reduction of oxygen [2]. The sheet resistance of rGO films decreases as the reduction temperature increases. This is attributed to an increase in the number of delocalized π bonds for sp^2 carbons by the reduction. The lower resistance should facilitate the separation and transportation of photo-carriers [3].

In this research, rGO was synthesized by the thermal decomposition of sucrose and reduced by thermal annealing under a nitrogen atmosphere [4]. A heterojunction diode was fabricated by depositing a rGO film on a cleaved n-type Silicon/Silicon Oxide (Si/SiO_2) substrate. We studied the response of the heterojunction to UV irradiation at room temperature. We will discuss the preparation and characterization of rGO, the heterojunction fabrication, and analysis of UV photoconduction.

Methods

rGO

Graphene oxide (GO) was prepared by hydrothermal carbonization of a 0.1 M solution of sucrose (sugar) in H_20 at 200°C. The GO was reduced by thermal annealing at 600°C for 2 h in a nitrogen atmosphere. The resultant rGO flakes were characterized using Scanning Electron Microscopy (SEM), Energy Dispersive Spectroscopy (EDS), profilometry, and UV-Vis spectroscopy. A schematic of the preparation process is depicted in Fig. 1.

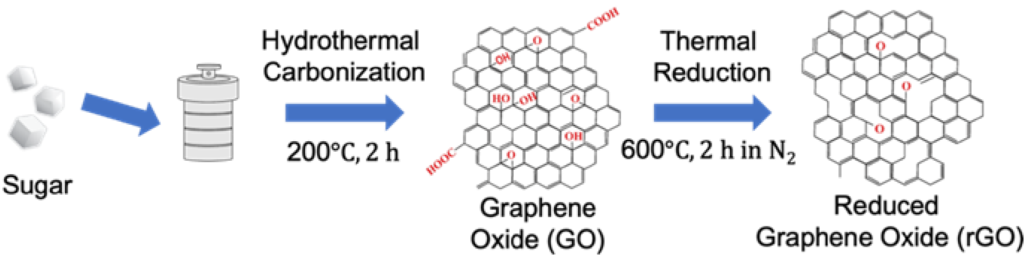

Fig.1. Schematic of rGO preparation process: GO is produced by hydrothermal carbonization of sucrose (sugar) and reduced to rGO by sintering under nitrogen.

Device

The rGO/n-Si diode was fabricated by using a commercially available n-doped Si wafer with a 200 nm oxide layer, with pre-patterned gold electrodes over the oxide surface. A Si/SiO_2 substrate was cleaved through the electrodes. The exposed cleaved surface had the edge of the gold electrode separated from the doped Si by the insulating SiO_2 layer. We then transferred the rGO film onto the edge of the substrate, bridging the insulating layer. The sample was oven dried in air at 70°C for at least 1 h before performing any measurements. A schematic of the device is shown in the inset of Fig. 3a.

UV-Photoconduction

The current-voltage (I-V) characteristics of the diode were measured in vacuum at room temperature using a Keithley 6517B electrometer by applying voltages from – 1 V to +1 V in darkness and after irradiation. A 4 W-UV-lamp (2.46 W/m^2) with a 365 nm wavelength, was used to irradiate the rGO-n-Si heterojunction. The measurements were conducted in the reverse bias region with no applied voltage (0 V) and low voltages of -0.1 V and -0.5 V. The times of exposure to UV light ranged from 5 to 20 s.

Results

rGO Characterization

Figure 2(a) shows a typical EDS spectrum and SEM image (inset) for a rGO film. The spectrum reveals 86.6 atomic % of Carbon and 13.4 % of Oxygen The average thickness of rGO films analyzed with profilometry (results not shown) was 110 nm. The optical bandgap of rGO was estimated from the UV-Vis absorption spectra in Fig. 2(b) (inset). The intercept of the best fit of the Tauc plot: $(\alpha h\nu)^2$ versus hν Fig. 2(b) gives a bandgap of $E_g = 2.12 \pm 0.01\ eV$ [5]. This value is consistent with others reported in literature that range between 1.00 and 3.11 eV [5-6].

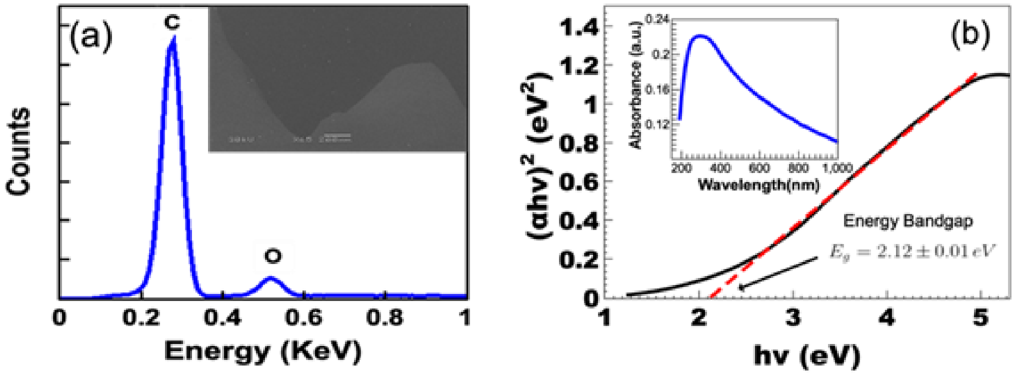

Fig. 2. (a) EDS spectrum and SEM image (inset) of rGO film.(b) Tauc plot with bandgap determination and UV-Vis spectrum (inset).

Heterojunction diode

The I-V plot of the rGO/n-Si diode at room temperature in darkness is shown in Fig. 3(a), with schematic diagrams of the device as insets. The results show the typical exponential behavior of a rectifying diode in forward and reverse bias. The diode current can be represented $I(T,V) = I_S\left(e^{\frac{qV}{nkT}} - 1\right)$, where I_S is the saturation current, q is the electron charge, V is the applied voltage, n is the ideality factor, k is the Boltzmann constant, and T is the temperature.

To study the photodetection properties and the rectification behavior of the device under irradiation we analyzed the photocurrent: $I_{ph} = |I_L - I_d|$ *and* responsivity: $R_p = \frac{I_{ph}}{PxA}$ [7-8]. In these equations, I_L is the current under irradiation and I_d is the current in darkness, P is the power density of the UV lamp, and A is the sample area impacted by light. The device maintains the rectification properties under UV illumination with a 365 nm wavelength. The values of the responsivity are depicted in Fig. 3(b). The high values of R_p suggest the phenomenon of carrier-multiplication or internal gain exist [7]. These results are consistent with others reported in literature for rGO heterojunctions. A heterojunction with other semiconductor material creates a built-in electric field at the junction that facilitates the separation of charge carriers. Prevention of electron-hole recombination and their effective collection make heterojunction photodetectors highly efficient [8].

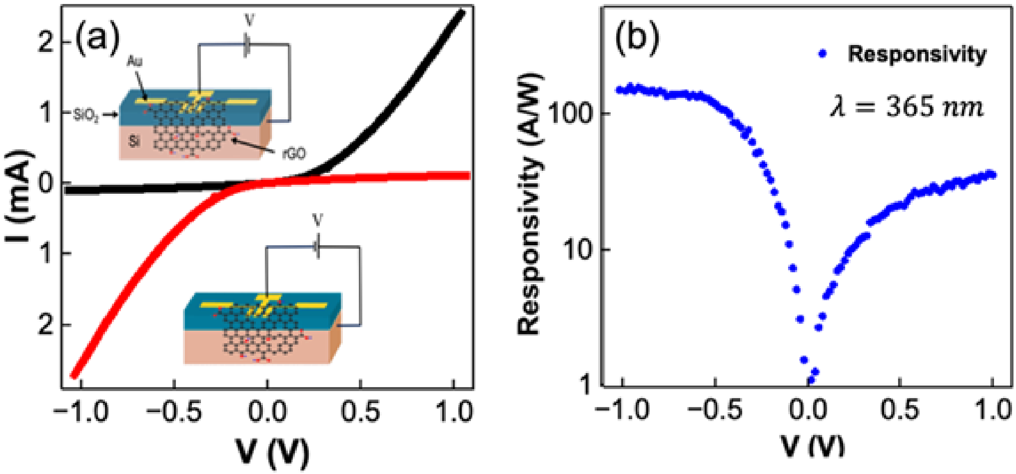

Fig. 3. (a) I-V characteristics of rGO/n-Si heterojunction in forward (black) and reverse (red) bias. The inset shows schematics of the device with electrical connections in forward and reverse bias. (b) Responsivity after irradiation with UV light for 5 s.

Figure 4(a) shows the optical response of the device for ON and OFF switching of the light, with applied voltages of -0.1 V and -0.5 V for time intervals of 5 s. The response and recovery times for -0.1 V and -0.5 V are shown in Fig. 4(b) and 4(c), respectively. The response time (t_{resp}) to UV light is computed as the time needed for the sensor to reach between 10% until 90% of the maximum current. The recovery time (t_{rec}) is the time to go from 90% to 10% of the maximum current. Various consecutive cycles with sharp transitions between the ON and OFF states were measured for our device. The measurements are stable and reversible. The response times are 1.2 s for -0.1 V and 1.4 s for -0.5 V. The recovery times are 1.6 s for -0.1 V and 1.7 s for -0.5 V. The values reported in literature for rGO devices vary considerably and range from microseconds to hundreds of seconds [7-8]. Fig. 4(d) shows the photosensor response to UV light without an external voltage source. The stable and reversible photoresponse suggest the rGO/n-Si heterojunction has the potential to be used in self powered devices.

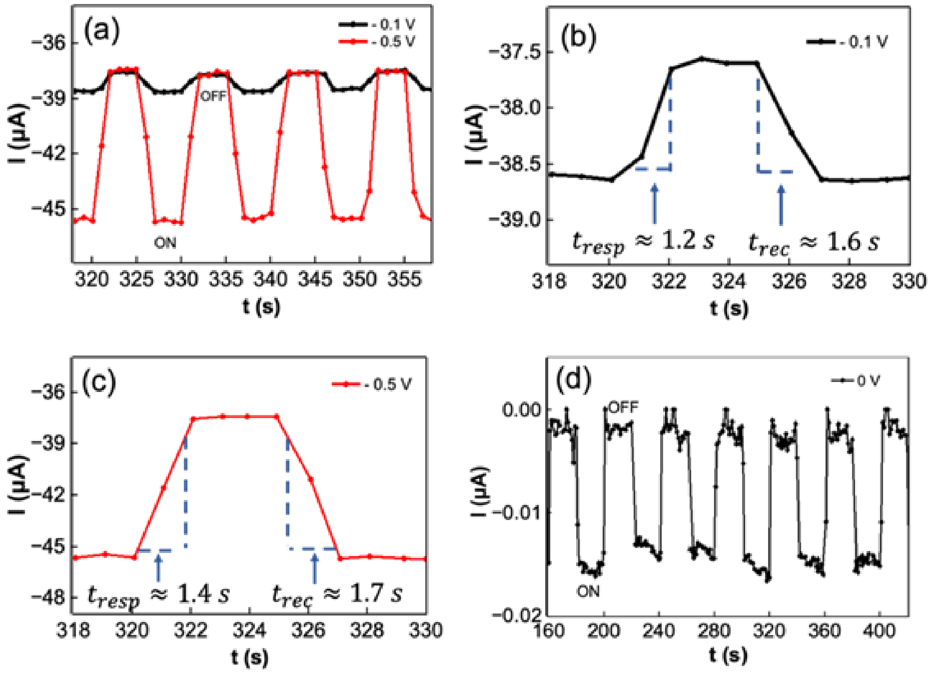

Fig. 4. (a) on-off response to uv light (365 nm) exposure in 5 s intervals, for applied voltages of -0.1 v and -0.5 v. (b) and (c) response and recovery times for a typical on-off cycle, with -0.1 v and -0.5v. (d) on-off response in 20 s intervals with 0 v bias.

Conclusions

High quality and electrically conductive rGO films were synthesized from hydrothermal carbonization of sucrose and thermal annealing. A rGO-nSi heterojunction was fabricated by depositing rGO onto the cleaved edge of a SiO_2/Si substrate. The I-V characterization of the device shows the typical behavior of a rectifying diode in forward and reverse bias. When exposed to UV light of 365 nm, the heterojunction shows good photoconduction properties with applied low voltages of -0.1 V and -0.5 V. The responsivity values of over 100 A/W suggest very good photodetection properties. The optical response of the device for the ON/OFF switching of the UV light, for the applied biases of -0.1 V, and -0.5 V, show stable cycles with response and recovery times between 1 and 2 s. The response to UV irradiation with 0 V is a promising result and show independence from the voltage source. The zero or low voltage photodetection properties, and the simple and inexpensive methods used to produce rGO and fabricate this device; make it attractive for the development of self-powered photoconduction devices.

References.

[1] Smith, A.T., LaChance, A.M., Zeng, S., Liu, B., Sun, L., Synthesis, properties and applications of graphene oxide/reduced graphene oxide and their nanocomposites, Nano Mater. Sci. 1, 31-47 (2019).

[2] Punckt, C., Muckel, F., Wolff, S., Aksay, I.A., Chavarin, C.A., Bacher, G., Mertin, W. The effect of degree of reduction on the electrical properties of functionalized graphene sheets, *Appl. Phys. Lett*. 102, 023114 (2013).

[3] Zhu, M., Li, X., Guo, Y., Li, X., Sun, P., Zang, X., Wang, K., Zhong, M., Wu, D., Zhu, H., Vertical junction photodetectors based on reduced graphene oxide/silicon Schottky diodes, *Nanoscale,* 2014,6, 4909-4914 (2014).

[4] Tang, L., Li, X., Ji, R., Teng, K.S., Tai, G., Ye, J., Wei, C., and Lau, S.P. Bottom-up synthesis of large-scale graphene oxide nanosheets, *J. Mater. Chem*. 22, 5676 (2012).

[5] Jilani, A., Othman, M.H.D., Ansari, M.O., Kumar, R., Alshahrie, A., Ismail, A.F., Khan, I.U., Sajith, V.K., Barakat, M.A., Facile spectroscopic approach to obtain the optoelectronic properties of few-layered graphene oxide thin films and their role in photocatalyst, New J. Chem. **41**, 14217-14227 (2017).

[6] Abid, P.S., Islam, S.S., Mishra, P., Ahmad, S. Reduced graphene oxide (rGO) based wideband optical sensor and the role of Temperature, Defect States and Quantum Efficiency, *Sci. Rep*. 8, 3537 (2018).

[7] Kumar, G., Prakash, N., Singh, M., Chakravorty, A., Kabiraj, D., Singh, S.P., Prabir, P., Khanna, S.P. Solution-Processed-2D on 3D Heterojunction UV-Visible Photodetector for Low-Light Applications, *ACS Appl. Electron. Mater*. 1, 1489-1497 (2019).

[8] Li, G., Liu, L., Wu, G. Chen, W., Qin, S., Wang, Y., Zhang, T. Self-Powered UV-Near Infrared Photodetector Based on Reduced Graphene Oxide n-Si Heterojunction, Small 12(36), 5019-5026 (2016).

Acknowledgments. This research is sponsored by the PENN-UPR Partnership for Research and Education in Materials (PREM) with funds from the USA National Science Foundation (NSF) Grant DMR-2122102.