

GRAPHENE-DERIVED BROADBAND PHOTODETECTORS FOR Si-COMPATIBLE OPTICAL SENSING APPLICATIONS

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Abstract

Graphene and its derivatives, particularly reduced graphene oxide (rGO), have attracted significant attention for broadband photodetection owing to their unique optoelectronic properties, solution processability, and compatibility with silicon (Si) platforms. The current paper reports the fabrication and characterization of rGO-based photodetectors designed for Si-compatible optical sensing applications. rGO was synthesized using a modified Hummers' method and characterized via Raman spectroscopy, X-ray diffraction (XRD), scanning electron microscopy (SEM), and thermogravimetric analysis (TGA) to confirm its structural integrity, reduction level, surface morphology, and thermal stability. The photodetectors exhibited a broad spectral response from the visible to near-infrared (NIR) region, with responsivity strongly dependent on the rGO layer thickness and applied bias voltage. The optimized devices demonstrated a peak responsivity of 0.5 A/W at 650 nm under a 5 V bias for a 3 μ L rGO layer, with fast switching characteristics and reasonable stability. Lower concentrations (1-2 μ L) exhibited reduced dark current and enhanced reliability, thus achieving a responsivity of up to 0.2 A/W. These devices function efficiently at low optical power levels ($\sim$ 30 μ W) and low voltages, making them suitable for integration into energy-constrained environments and CMOS-based systems. The proposed rGO-based photodetectors offer a cost-effective and scalable solution for broadband low-power optical sensing, highlighting their potential in biomedical diagnostics, environmental monitoring, and silicon photonic circuits.

Keywords: Broadband photodetector; Reduced graphene oxide; Responsivity; Optical sensing; Optoelectronic

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Introduction

Photodetectors are key components in many technological applications because they convert light into electrical signals by facilitating the interaction between the light and a material (Qiu *et al.*, 2017). They have shown excellent broadband detection properties, such as sensitivity, internal and external quantum efficiency, response speed, adaptability, and detection bandwidth. Therefore, photodetectors are promising candidates for sensing, imaging, and environmental monitoring applications (Pospischil *et al.*, 2013; Chiang *et al.*, 2015). In this scenario, the performance of the sensor is significantly influenced by dimensionality, and the inadequate ability of the material selected for the broadband photodetector to detect UV radiation could be a key limitation (Abbas *et al.*, 2024). Wideband photodetectors based on semiconductor materials such as GaN, SiC, ZnO, and TiO are presently considered to be the best candidates for UV light detection, but their range of applications is constrained by their insensitivity to visible light (Zou *et al.*, 2018; Fu *et al.*, 2019; Liao, 2021). A rapid pace of aging, as well as several ailments, including skin cancer and cataracts, have been connected to excessive UV exposure (Kumar *et al.*, 2021). Other potential adverse effects are 1% reduction in ozone layer thickness, an increase in UV radiation by 2%, and rise in melanoma indices by 3% (Leckner, 1978; Ziegelberger *et al.*, 2006). Additionally, silicon is a frequently used material for photodetection owing to its reliability, low bandgap, naturally occurring production, and surface states; however, it is not widely used in the UV to IR region (Ajiki *et al.*, 2016; Wang *et al.*, 2016). Figure 1 shows the steps involved in the synthesis process of GO and rGO.

Silicon consists of a low penetration depth of light, resulting in a photo responsivity of < 0.1 A/W and a high reflection coefficient for the near UV range (Kuschnerus *et al.*, 1998). Recent developments in nanotechnology have provided breakthroughs and novel routes for researchers to explore innovative materials for a broad range of applications. Broadband photodetection requires a high-speed ultra-surface junction that effectively

separates and collects charges. Graphene has become one of the most renowned breakthroughs in the post-silicon age because of its unique and fascinating properties, that is, a broad absorption spectrum (Gabor *et al.*, 2011, Fu *et al.*, 2023), very low mass, excellent conductivity, large surface to volume ratio, and high charge mobility (Ferrari *et al.*, 2014, Barde & Waghuley, 2013). Its strong π - π band structure implies that it is a semi-metallic substance.

The conduction and valence bands exhibit symmetry at the Dirac point (charge neutrality point). Their unique electrical and optical attributes originate from the Dirac massless electrons dispersed linearly at near-light speeds (Abid *et al.*, 2018). Pure graphene has a Fermi energy near the charge-neutrality point; hence, its electrical characteristics around the Dirac point (k-point) should have the lowest conductance. Graphene has zero bandgap energy, which causes a very short life span of carriers and poor responsivity, though its low absorbing capacity of 2.3% of incident light with high mobility and ultra-fast detection makes it more suitable for photodetection applications (Bonavolontà *et al.*, 2021). Defects can be introduced to create mid-gap states in the band structure of pristine graphene (Gan *et al.*, 2013). Graphene is an extremely useful material for Fin field-effect transistors (FinFETs). Graphene oxide (GO) and reduced graphene oxide (rGO) are two main promising candidates to resolve the issue because the bandgap of GO is 2.2 eV and that of rGO varies from 1 to 1.69 eV (Abid *et al.*, 2018). Jin *et al.* reported tuning the band gap of rGO from 0.264 to 0.786 eV by controlling the concentration of oxygen-containing groups using oxidation treatment with nitric acid (Jin *et al.* 2020; Khairudin *et al.*, 2024). The bandgap of rGO can be tuned by managing the removal of oxygen functional groups during the reduction process of GO to achieve excellent conductivity and optical transmittance (Chang *et al.*, 2013; Tielrooij *et al.*, 2013). Consequently, significant efforts have been made to improve the synthesis process for enhancing the liquid-phase exfoliation during prolonged

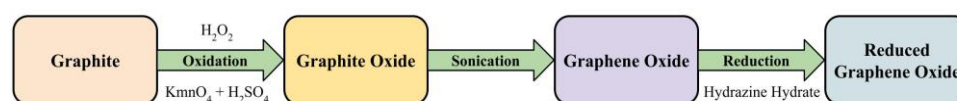


Figure 1. Steps of synthesis process of GO and rGO

sonication. However, large-scale manufacturing remains unfeasible. Researchers have been striving to address this dilemma by considering several factors, including the synthesis, device architecture, and fabrication methods. To optimize the manufacturing process, work has been carried out on the absorption area, generation of excess electron-hole pairs, and minimization of recombination to enhance the photo response by adding band defects of rGO. Currently, the most popular methods used to synthesize GO and rGO are the Brodie, Hummer, and Staudnmaier methods (Hummers & Offeman, 1958; Khan *et al.*, 2021; Mattson *et al.*, 2013). In this paper, we report a photodetector based on rGO to detect visible to infrared wavelengths in the 450–800 nm range. The morphological and electrical characteristics of GO and rGO were analyzed using Raman spectroscopy at 532 nm, XRD, FESEM, FTIR, AFM, and TGA. These properties indicate that the synthesized GO and rGO are promising for visible and infrared light-sensing applications. Further, the light detection response of the proposed photodetector for 1, 2 and 3 μL rGO thin films was determined by varying the voltage, power density, and excitation wavelength. The extremely sharp ON-OFF switching characteristics to illumination of light on the surface of the proposed photodetector (PD) have been obtained for a wide range, that is, visible to IR (450–800 nm) for 1–5 volt and low power density. The highest responsivity 0.2 A/W and fast response time of 0.08 were achieved for 550 nm wavelength at a power intensity of 30 μW .

Experimental Setup

Hummer's processes (Eda *et al.*, 2008; Zhu *et al.*, 2010) have been adapted to produce GO from graphite flakes as depicted in Figure 1. Initially, 2 gm of graphite and 50 mL concentrated sulfuric acid were mixed. A suspension was obtained after continuous stirring and cooling in ice water. Six grams of the oxidizing agent KMnO_4 were progressively applied to the solution at a temperature below 10°C . After stirring at room temperature for 25 min, the suspension was sonicated for 15 min using an ultrasonic bath. The stirring-sonication process was repeated 10 times after adding 250 mL of distilled water to the mixture. Subsequently, 30 mL of hydrogen peroxide (H_2O_2) was added to the exfoliated graphite oxide, which was accomplished by ultrasonically agitating the scattered solution for an hour. After centrifugation at 10000 rpm, GO was cleaned several times with HCl and distilled water (to remove extra metallic ions) until the pH

reached 7. The GO precipitate was then allowed to dry at room temperature for 24 h. The suspension was sonicated for 2 h to obtain the rGO. Hydrazine hydrate was mixed dropwise into an exfoliated graphite oxide suspension at room temperature. For this sample, GO and hydrazine hydrate were maintained at a weight ratio of 7:9. The reduction was performed at 100°C for an hour. After passing the black precipitate through cellulose filter paper to obtain a neutral pH, it was rinsed with distilled water and a 1M HCl solution. The filtrate was allowed to dry at room temperature for a full day to extract rGO. Figure 2 illustrates the detailed configuration of the experimental setup employed for conducting optical electrical tests on the rGO photodetector.



Figure 2. Experimental steps for fabrication and testing of rGO photodetector

Morphological/Structural Analysis

The structural changes and topography were determined using morphological analysis, which was carried out using SEM for both rGO and GO photodetectors. The removal of oxygen functional groups in rGO likely resulted in changes in surface morphology (Ashery *et al.*, 2021). Figure 3(a) and 3(b) clearly illustrate the layered and flaky microscopic structures of rGO and GO, respectively. A comparatively close surface pattern was observed in the rGO composites. The exfoliation of the GO layers during ultrasonic treatment is illustrated in Figure 3(b). The crystallized layered microstructure is clearly visible in the images. The typical RMS roughness and peak-to-valley height for GO, as measured by AFM, are 7 nm, 5.34 nm, and 48.7 nm, respectively.

Figure 3(a) illustrates that the interlayer spacing is greater and the edges are thinner in GO compared to in (rGO Raman spectroscopy provides insight into the inelastic scattering (Raman) of a molecule during the bombardment of monochromatic light. Raman spectroscopy was used to track the structural changes in a sample as it underwent oxidation and reduction. The Raman spectra of GO and rGO at 532 nm are shown in Figure 4(a). Raman spectra were recorded at a laser power of 35 mW. The spectra reveal two characteristic bands at 1333–1365 cm^{-1} for the D-band (disorder) and at 1568–1579 cm^{-1} for the G-band (graphite). As shown in Figure 4(a), two fundamental vibrations observed in the range of 1100–1700 cm^{-1} for rGO and GO. The breathing mode of the J-point phonons with A_{1g} symmetry forms the D band, which can be seen at 1347 cm^{-1} and 1355 cm^{-1} for rGO and GO, respectively.

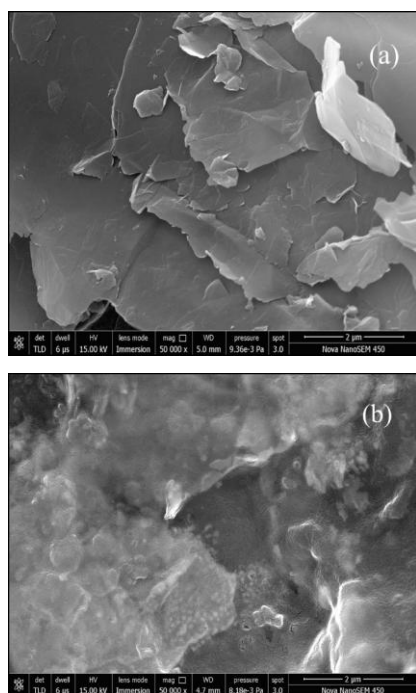


Figure 3. FESEM Images of (a) GO and (b) rGO

In contrast, the G band vibration appeared at 1579 cm^{-1} for rGO and 1598 cm^{-1} for GO. The stretching C–C bond, which is present in all sp^2 carbon structures, also contributes to the second band, G. The I_D/I_G has been shown to be 0.92 and 0.37 for GO and rGO, respectively. Double resonance transitions, which result in the creation of two phonons with opposite momenta, are thought to be responsible for the 2D peak. The lower intensity and broadness of the 2D peaks also indicate disorder

in GO. XRD analysis was performed to measure the crystal phase and investigate the interlayer spacing of the GO and rGO. Figure 4(b) shows the XRD patterns of rGO and GO to determine whether the material is crystalline or amorphous.

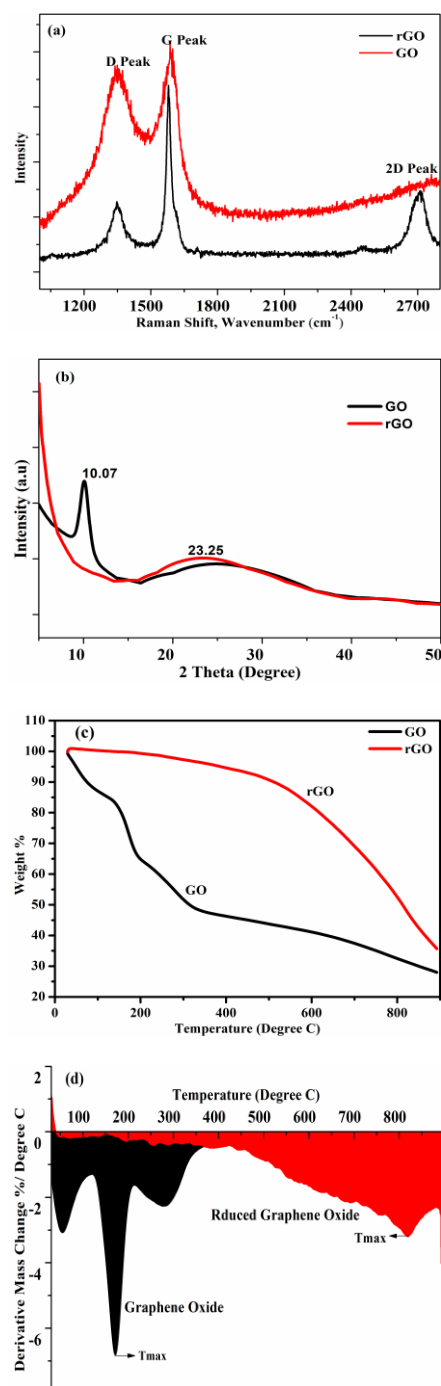


Figure 4. (a) Raman spectra of GO and rGO at 532 nm. (b) XRD pattern of GO and rGO. (c–d) Thermo gravimetric analysis (TGA) as function of temperature

$$n\lambda = 2d \sin \theta \quad (1)$$

where λ is 1.5402 Å (wavelength of the incident X-ray), d is the interplanar spacing in Å, n is the order of diffraction (1), and θ is the peak position. Reflection peaks of the XRD patterns of GO and rGO were observed at $2\theta = 10.07^\circ$ and 23.25° respectively. The sharp peak of GO indicated a highly oxidized state in the synthesized GO sample. The whole product was oxidized completely at a peak value of 10.07° with a d -spacing 8.77 Å. The water molecules and binding of oxygen-containing groups (epoxy, carbonyl, carboxyl, and hydroxyl groups) between the layers of graphite cause an interlayer gap between consecutive carbon basal planes. The reduction of GO was validated by XRD examination, with a wide diffraction peak at $2\theta = 23.25^\circ$ for rGO, corresponding to a d spacing 3.82 Å. This indicates that the π -conjugated arrangement of graphene was significantly restored in the synthesized rGO. The shift and expansion of the peak correspond to the short-range order of the very thin layers of rGO caused by a higher degree of exfoliation. The peak at 10.07° vanished after the chemical reduction of GO, whereas the intensity of the rGO peak at 23.25° increased with increasing reduction time. The reduction of the oxygen functional groups caused a reduction in the d -spacing, confirming the conversion of GO into rGO. The overlapping of short-range ordered layers of rGO sheets causes the strength of the diffraction peak to increase with reduction time.

It is important to control the quality of GO and rGO. To investigate the thermal stability, the Parkin Elmer method was used to measure the significant loss in the mass of rGO and GO. Weight loss was observed in the temperature range of 0-1000°C under a nitrogen atmosphere. The collected data shown in Figure 4 (c-d) clearly demonstrate the mass loss in both the GO and rGO samples. In GO, substantial loss in weight is observed up to 95°C (12%), 150°C to 200°C (21%), and 200 to 340°C (16%) due to water elimination and removal of oxygen functional groups. A small weight loss was observed in rGO compared to GO.

$T_{\max}$ observed from TGA carbon combustion peaks was at 170 °C and 825 °C, respectively. The carbon combustion peaks increased as the particle size increased. GO has a lower $T_{\max}$ than rGO so it consumes less heat energy to overcome the weaker non-graphitic sp^3 hybridized carbon with a large density of defects following the severe oxidation reaction. GO also has a high concentration of

oxygen groups, which contribute to the lower energy required for decomposition. $T_{\max}$ of rGO is higher because the covalent bonds of sp^2 hybridized carbon atoms require more heat energy. $T_{\max}$ is a key characteristic for detecting the presence of graphite and GO impurities in graphene.

The responsivity and response time were measured for three different thin films (volumes, 1, 2, and 3 μL) fabricated using the drop casting process. Furthermore, the dependence of the photocurrent was measured by tuning the excitation wavelength and the applied voltage.

Results and Discussion

A silicon substrate-based interdigitated electrode (IDE) was fabricated using a photolithographic process. A photodetector with rGO on a silicon-based substrate and gold connections was designed to detect a wide optical range of 450-800 nm. A thin film of rGO with volumes of 1, 2 and 3 μL was deposited on a silicon substrate via drop casting, as shown in Figure 2. Gold connections were made between the rGO photodetector and the external circuit to ensure significant electrical signal transmission.

The oxygen group elements decreased in the rGO sample owing to the reduction in GO, thereby increasing electron photogeneration. 2-D material thin film photodetectors have shown a strong matter–light interaction. This interaction results in a quick ON-OFF switching owing to excessive absorption. To assess the optical sensing performance, the designed photodetector was exposed to visible to infrared light, which causes a sharp increase in the current peaks, particularly when it changes from dark to a specific intensity. In terms of the response time and responsivity, we achieved better results for illumination at wavelengths between 450 and 800 nm. Moreover, the dependencies of the exposed wavelength, applied voltage, and rGO volume were also demonstrated.

Figure 5 illustrates the current-voltage characteristic curves of the drop-cast GO and rGO thin films. The linear characteristics of rGO were more pronounced than those of GO because of the reduction of oxygen, making it advantageous for photodetection applications. Conductivity was found to improve with fewer oxygen-containing functional groups. GO has insulating properties owing to its enormous amount of oxygen functional groups.

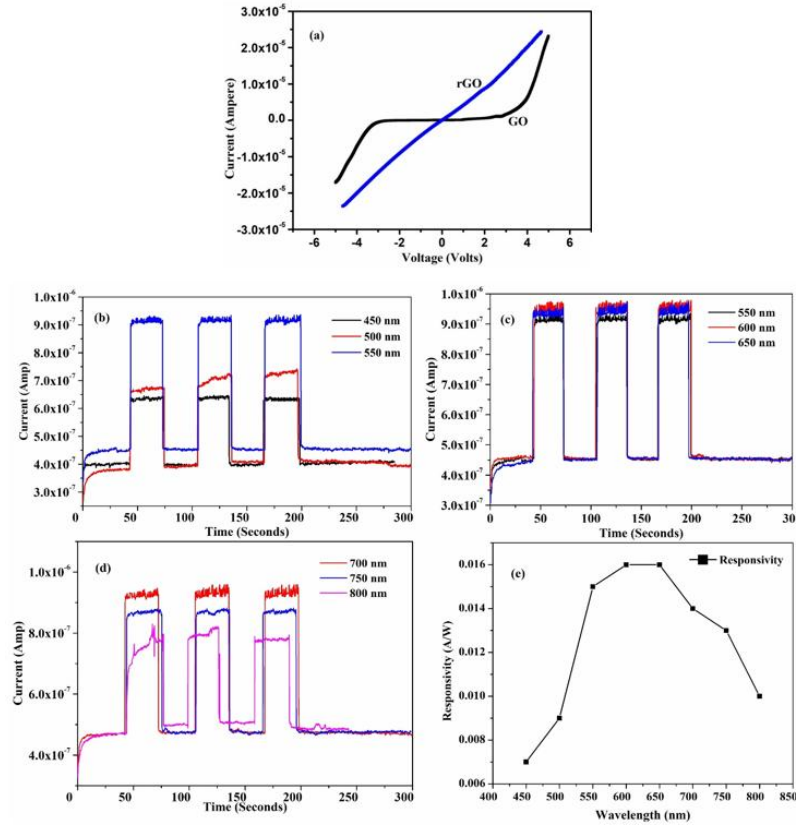


Figure 5. (a) I-V characteristics of GO and rGO. The time vs current on - off switching characteristic of rGO photodetector depending on wavelength (b) 450-550 nm (c) 550-650 nm and (d) 700-800 nm (e) Corresponding responsivity

A thin film of rGO was prepared by drop-casting at a concentration of 1 μL for the initial setup shown in Figure 2. A photocurrent of 0.632 μA was detected when exposed to 450 nm light, suggesting a robust light-material interaction. The photo current increased to 0.658 μA (4.1% improvement) when illuminated with 500 nm light. The highest photo response value of 0.953 μA was achieved at 600 nm with an applied voltage of 1 volt and power of 30 μW , suggesting efficient photon absorption and interband transition in rGO. The generated current exhibited better stability at 550 nm, with a value of 0.913 μA . Moving into the visible light range (450–600 nm), the peak current of the proposed rGO photodetector increased by 47.18%. However, the current peaks decreased by up to 13% when illuminated with light in the wavelength range of 650 to 800 nm, although the responsivity and response time remained satisfactory in this region. As shown in Figure 5, the sensor demonstrated excellent repeatability and stability in its response, with the responsivity being a measure of the device's ability to convert photonic energy into an electrical signal. The responsivity

and sensitivity of the rGO photodetector were also measured, using the following equations.

$$\text{Responsivity } (R) = (I_L - I_D) / P_o \quad (2)$$

$$\text{Photosensitivity} = (I_L - I_D) / I_L * 100 \quad (3)$$

where I_L and I_D denote the currents under illumination during light and in the dark, respectively, and P_o is the power density. The wavelength-dependent responsivity lies between 0.007 A/W and 0.016 A/W over a wide range of wavelengths, from 450 nm to 800 nm. The maximum response of PD has been observed at 550 nm, 600 nm, 650 nm, and 700 nm optical ranges, e.g., 0.015 A/W, 0.016 A/W, 0.016 A/W, and 0.014 A/W, respectively, with 51% photosensitivity for an ultrathin (1 μL) rGO layer at 1-volt biasing voltage and 30 μW power. The rising time (τ_r) and fall time (τ_f) are important performance parameters that indicate the switching characteristics of a photodetector. It is observed that response time is continuously improved and shows better sensitivity with increase in wavelength range

450 nm-800 nm from 0.37 seconds to 0.07 seconds, respectively. Repeatability was assessed by monitoring the photocurrent response while exposing the device to several consecutive cycles of light on and off. Good operational stability and dependable photo response behavior of the rGO photodetector are indicated by the constant amplitude and form of the photocurrent signals throughout multiple cycles. This proves that short-term cyclic operation does not substantially impair the device's performance, which is a crucial feature for real-world applications.

In the preceding section, the evaluation and reporting of the photocurrent as a function of wavelength were conducted. Further, the analysis of the voltage dependency of the realized rGO device in terms of photocurrent and responsivity is observed for thin film volumes of 1, 2 and 3 μL at 550 nm and 30 μW . Variations in the voltage from 1 V to 5 volts linearly increase the corresponding photocurrent, as depicted in Figure 6. The maximum responsivity and fast response time of the photodetector obtained for the sample of 1 μL is 0.19 A/W and 0.08 seconds, 2 μL is 0.2 A/W and 0.08 seconds at 5 volts biasing voltage. Higher voltages enhance the electric field across the photodetector, boosting the generation and separation of charge carriers. Figure 6 (c) shows that the peak responsivity value reaches 0.5 A/W;

however, there is instability in the photocurrent for a 3 μL rGO drop. This effect is more significant because of the increase in dark current produced at higher voltages. Owing to structural disorder and residual oxygen functions, hopping conduction between localized states typically dominates charge transfer in thinner rGO films. Improved penetration channels and decreased sheet resistance can increase carrier mobility as the thickness increases. However, because of the longer carrier diffusion routes and potential trapping at the defect sites, thicker films also result in higher recombination. When determining the ideal film thickness to provide high responsivity and quick reaction, this trade-off is essential. To avoid any adverse thermal effects, the obtained data may serve as a basis for using a broadband photodetector device. The proposed photodetector produces 0.2 A/W responsivity with minimum response time about 0.08 seconds which is well suited as a photo sensor application in the optical range at 550 nm. The above results show that the rGO-based photodetector can detect radiation from visible to IR wavelengths and that it particularly improves the photoresponsivity, response time, and sensitivity at very low power intensity and applied voltage. The results were compared with those of other photodetectors, as shown in Table 1.

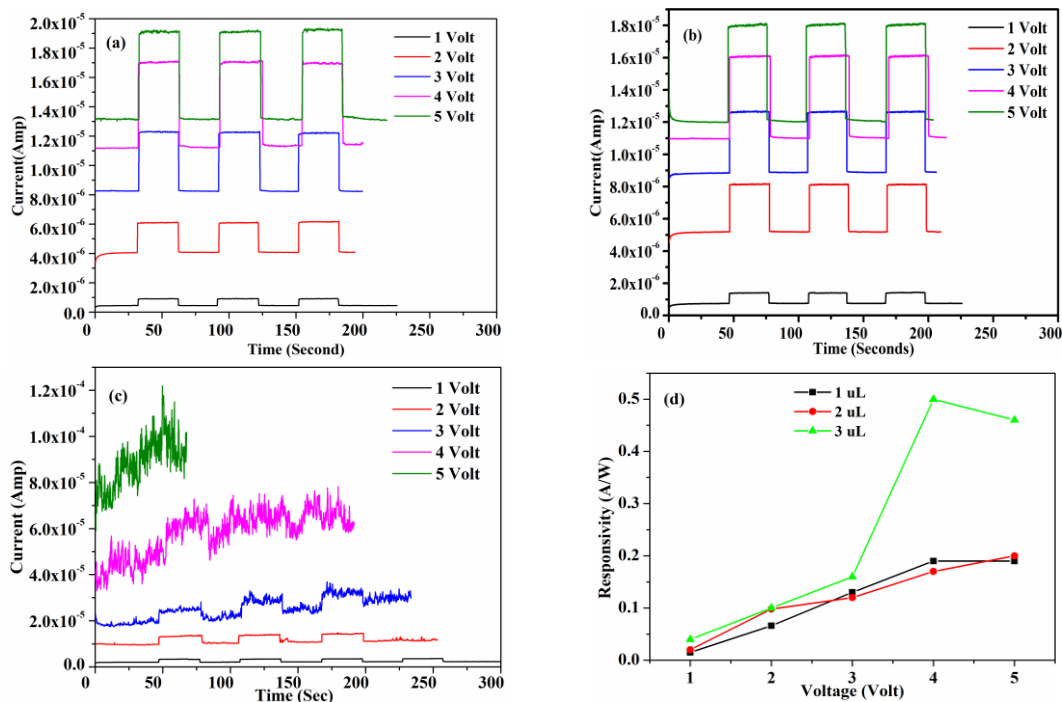


Figure 5. (a - c) Voltage dependent Current response of rGO photodetector as a function of time at 550 nm. (d) Corresponding responsivity at different voltage

Table 1. Performance matrices for ultra violet to Infrared photodetectors based on GO and rGO

Device structure	Responsivity (A/W)	Photo-sensitivity	Response time (Seconds)	Response range	References
GO junction	0.0236	-	0.13	290-1610 nm	Lai <i>et al.</i> 2014
rGO/n-Si heterojunction	0.20	81.2	-	370-815 nm	Bonavolontà <i>et al.</i> 2021
rGO	0.00012	49.2%	1.5 and 1.7	635-1064 nm	Sehrawat <i>et al.</i> 2018
GO/SiNW	0.009	-	-	532 nm-118.8 μ m	Cao <i>et al.</i> 2014
rGO	0.73	-	3.4 and 5.2	465-625 nm	Alsaedi <i>et al.</i> 2017
rGO	0.0016	-	114 *10 ⁻⁶ and 678 * 10 ⁻⁶	974 nm	Ahmad <i>et al.</i> 2018
rGO	0.20	33%	0.08 and 0.08	450-800 nm	Present Work

Conclusions

In this study, we demonstrate the regulated photo response of rGO-based photodetectors operating in the visible to infrared spectral range. GO and rGO were synthesized using the Hummers method, and their structural, morphological, and thermal characteristics were systematically analyzed using Raman spectroscopy, XRD, SEM, and TGA. These characterizations confirm the successful reduction of GO and the formation of rGO with favorable properties for enhanced photodetection. The photodetector performance was evaluated for different rGO layer thicknesses (1, 2 and 3 μ L), revealing wavelength and bias dependent photoresponsivity. A maximum responsivity of 0.016 A/W with a fast response time of 0.08 s was achieved at 600-650 nm under a 1 V bias and 30 μ W optical power for the 1 μ L rGO layer, accompanied by a sensitivity of 52%. At 550 nm, a stable photocurrent and responsivity of 0.015 A/W were observed, highlighting good device reliability. The responsivity increased to 0.19 A/W and 0.2 A/W for 1 μ L and 2 μ L rGO concentrations, respectively, under a 5 V bias, exhibiting sharp ON-OFF switching behavior. While the 3 μ L rGO layer showed a further enhancement in responsivity up to 0.5 A/W, it also exhibited increased dark current and reduced stability. A comparative analysis with existing photodetectors indicates that the developed rGO-based devices offer competitive performance at significantly lower power densities and voltage levels. The study results underscore the potential of ultrathin rGO films for low-power broadband photodetector applications, particularly where energy efficiency and compact form factors are critical.

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Author Contributions

Validation: R.K.M., R.M., S.K.; Formal Analysis: P.K.I.; Investigation: P.K.I.; Data Curation: P.K.I.; Writing – Original Draft: P.K.I.; Writing – Review & Editing: R.K.M., R.M., S.K.; Supervision: R.K.M., R.M., S.K. All authors have read and agreed to the published version of the manuscript.

Ethics Statement

Ethical approval was not required for this study, as it did not include human participants or animals, involve identifiable personal data, or use materials subject to institutional ethical review.

Declaration of Generative AI Use

During the preparation of this manuscript, the authors used Quillbot to improve the English language and flow of the manuscript. The tool was used solely for grammar and language refinement. All AI-assisted outputs were critically reviewed, verified, and revised by the authors, who accept full responsibility for the content of this manuscript.

Conflict of Interest

The authors declare that they have no conflicts of interest related to this work.

Data Availability Statement

No data were generated or analyzed in support of this research

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