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# Optical Sensor Fabricated From Graphene Oxide/$Ga_2O_3$ Composite

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**ABSTRACT**

Graphene oxide (GO) is a promising material for several applications. The studies of GO optical sensors have been conducted in numerous reports in recent years. Especially, producing GO from disposed zinc batteries has attracted much attention because it facilitates human to protect the environment. This study arms to produce GO/$Ga_2O_3$ composite to make optical sensors. The GO nanosheets were exfoliated from disposed graphite rods of waste batteries. $Ga_2O_3$ was then deposited on GO via hydrothermal method. The GO/$Ga_2O_3$ film was deposited on filter paper using vacuum filtration method for fabricating the optical device. A field emission scanning electron microscope (FESEM) image indicates that GO nanosheets exfoliated by electrochemical method are multi-layers. FESEM image also reveals that $Ga_2O_3$ was successfully deposited on GO. The optical sensor exposed the strong repeatability and reliability. The photocurrent of 9 µA measured at 10 V under the excitation of 650 nm light was obtained. The rise time and fall time of sensor are determined at 3.7 s and 4.2 s, respectively. The results indicate that waste zinc batteries can be used to produce valuable products.

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## 1. Introduction

Graphene oxide (GO) is a 2D material that has high potential applications in electronic and optoelectronic devices [1]. It has the band gap from 0.11 eV to 3.0 eV depending on densities of oxygen functional groups on its surface [2]. GO can be modified to reduced GO (rGO) using thermal, chemical, and photothermal methods [3]. The rGO has been reported to have several applications in energy storage, electronic devices, biosensors, electrochemical sensors, and optical sensors [4]-[8]. rGO has electron mobility of 300 $cm^2V^{-2}s^{-1}$ [9], significantly higher than common organic semiconductors (< 1 $cm^2V^{-2}s^{-1}$) [10]. Electrochemical is an environmentally friendly method to exfoliate GO because its electrolytes are not toxic as compared to those used in Hummer's methods. The electrochemical method has been adapted to recycle graphite rods from disposed zinc batteries [11], [12], opening opportunity to produce high valuable GO from hazardous waste.

Beta gallium oxide ($\beta$-$Ga_2O_3$) has obtained a lot of consideration for several applications in photocatalysts, solar cells, gas sensors, power devices, UV photodetectors, due to its ultrawide band gap of 4.8 eV, leading to a high critical field of 8 MV/cm [13], [14]. $\beta$-$Ga_2O_3$ reveals several advantages over other wide band materials such as SiC and GaN because high-quality and low-cost bulk substrates can be grown using low cost processes such as Czochralski (CZ) [15], floating zone (FZ), edge-defined film-fed (EFG) methods [16]. Current research based on $\beta$-$Ga_2O_3$ focuses mainly on power electronics and optoelectronic devices. Its potential applications in military communications and highly secure NLOS (Non-Line-of-Sight) have significantly increased its attention [17].

Although photodetectors have various applications in industry, its manufacturing based on traditional technologies requires high cost of materials and processes [18]. The usage of GO to fabricated optical sensors opens new era to produce high efficiency and low-cost photodetectors [18]. Response time and responsivity are two main factors to be used in determining the figure of merit of optical sensors. It was reported that the rGO optical sensor prepared by vacuum filtration has a responsivity of $1 \times 10^{-4}$ A/W and response time of 1.5 s under illuminating of 635 nm light source [19]. The integration of rGO on n-type Si substrate produces a light sensor with the responsivity of 0.2 A/W and response time of 1.9 s under excitation of 685 nm source [20]. J. An utilized laser to write patterns on rGO-ZnO composite films to attain a light sensor with the high responsivity of 3.24 A/W and the response time of 17.9 s under excitation of 365 nm light source [21]. The results indicated that the combination of rGO and wide band gap materials leads to an extremely high performance of optical sensor under the UV illumination.

This work aims to develop optical sensors based on GO exfoliated from disposed graphite rods of waste zinc batteries. GO was exfoliated using electrochemical methods. The GO powder was then mixed with $Ga(NO_3)_3$ and $NH_4NO_3$ solution in autoclave and annealed to obtain the composite of GO and $Ga_2O_3$. The composite powder was deposited on filter papers to form a thin film using vacuum filtration method. The optical sensor was then fabricated on GO/$Ga_2O_3$ composite films to take advantages of both GO and $Ga_2O_3$.

## 2. Experimental details

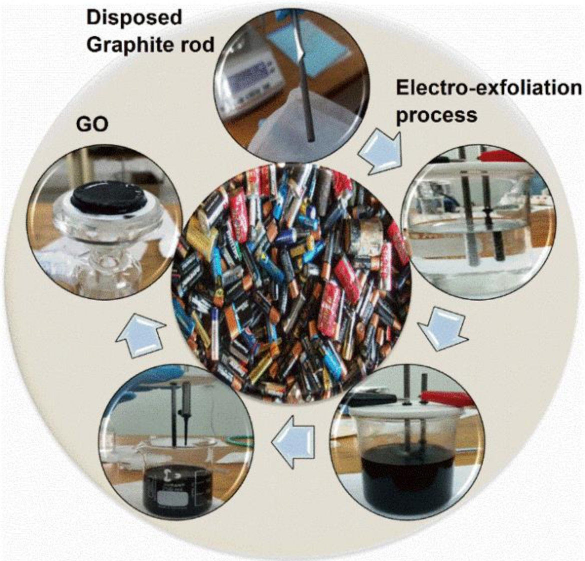


**Figure 1.** *Schematic illustration of the fabrication of GO from disposed graphite rods of zinc batteries.*


**Figure 2.** *Schematic of preparing GO/$Ga_2O_3$ composite using hydrothermal method.*

Figure 1 show the schematic of recycling GO from disposed zinc batteries. The process includes: (i) collecting disposed zinc batteries, (ii) detaching graphite rods, (iii) exfoliating GO using electrochemical method, (iv) filtering GO powder using filter paper. The electrolyte used in this study is $(NH_4)_2SO_4$ 2.5% (Scharlau, Spain) because it is relatively less toxic as compared to other solution [12]. Figure 2 show the schematic of fabricating GO/$Ga_2O_3$ from GO powder and $Ga(NO_3)_3$ (Wuhan Tuocai Technology, China). The fabrication process includes: (i) mixing GO, $Ga(NO_3)_3$, and $NH_4OH$ (PH of solution is kept at 9), (ii) sonicating solution in 30 minutes, (iii) stirring solution in 2 hours, (iv) putting solution in autoclave and annealing, (v) Obtaining solution after hydrothermal process, (vi) drying to gain $GO/Ga_2O_3$ composite powder.

Figure 3 reveals a procedure of fabricating $GO/Ga_2O_3$ optical sensor. The process includes: (i) a $GO/Ga_2O_3$ solution was prepared by mixing 3 mg $GO/Ga_2O_3$ with 1 ml DI, and the solution was sonicated to disperse $GO/Ga_2O_3$ in DI. (ii) 5 ml $GO/Ga_2O_3$ solution was dropped on the acetate cellulose filter (pore size of 0.22 μm) to form $GO/Ga_2O_3$ thin film. To form electrodes on $GO/Ga_2O_3$ surface, a shadow mask was employed, and Ti/W was deposited on $GO/Ga_2O_3$ film using sputtering method.

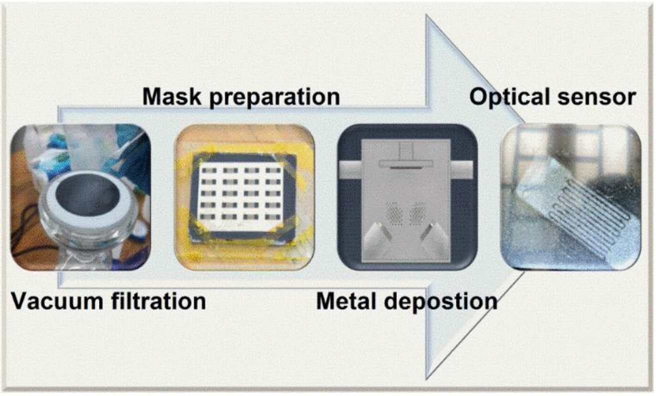


**Figure 3.** *Schematic of fabricating GO optical sensor from electrochemical exfoliation GO.*

## 3. Results and Discussion

Figure 4 (a) shows a field emission scanning electron microscope (FESEM) image of GO exfoliated by electrochemical method, revealing the stacking of GO nanosheets with some overlaps and wrinkles. The morphology of GO nanosheets exfoliated in $(NH_4)_2SO_4$ 2.5% is in agreement with those exfoliated in NaOH and KOH under the assistant of arc [12]. Figure 4(b) illustrates the FESEM image of $GO/Ga_2O_3$ composite. The percentage of $Ga_2O_3$ is 10%. The unification of $Ga_2O_3$-GO reveals that $Ga_2O_3$ nanoparticles tended to accumulate along the wrinkles and edges of GO.

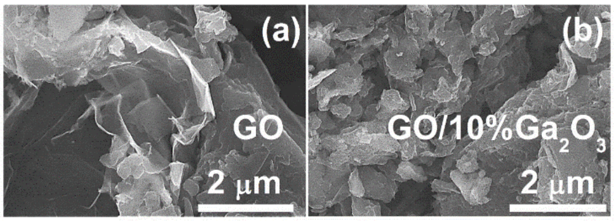


**Figure 4.** *FESEM images of (a) GO exfoliated in $(NH_4)_2SO_4$ 2.5% and (b) $GO/Ga_2O_3$ composite fabricated by hydrothermal method.*

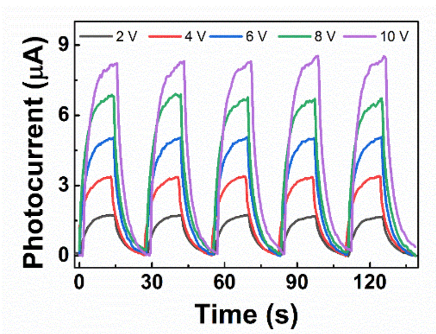


**Figure 5.** *Current – time characteristics for several cycles as light is turned on and off. The sensor was excited by 650 nm light. The voltage applied between two electrodes are varied from 2.0 V to 10.0 V, and the excitation power is 45 mW/cm$^2$.*

Figure 5 shows the current – time characteristics of resistive optical sensors fabricated from $GO/Ga_2O_3$ composite film. The photoresponse is measured at a constant intensity of 45 $mW/cm^2$ of incident light that has the wavelengths of 650 nm, corresponding to the excitation energy of 1.9 eV. The sensor is observed to be stable and repeatable in response to a photocurrent to be calculated followed formula:

$$I_{ph} = I_{light} - I_{dark}, \quad (1)$$

where $I_{ph}$, $I_{light}$, and $I_{dark}$ are photocurrent, light current, and dark current, respectively. The values of $I_{ph}$ as a function of time and voltage are illustrated in Fig. 6(a), showing that at bias voltages ranging from 2.0 V to 10.0 V, $I_{ph}$ significantly increases (~ 4 times) at excited wavelength of 650 nm. The relationship between photocurrent and applied voltage is shown in Fig. 6(b), illustrating a linear behavior of the photocurrent.

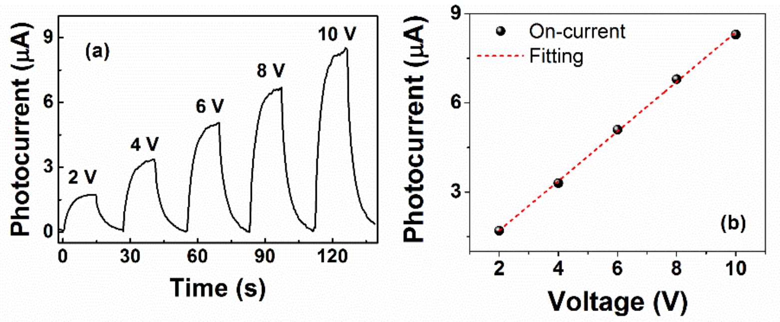


**Figure 6.** *(a) and (b) Comparison of the temporal photocurrent response of $GO/Ga_2O_3$ sensors at bias voltages ranging from 2.0 V to 10.0 V.*

Figure 7 shows the responsivity of $GO/Ga_2O_3$ composite sensor, which is calculated from [22]:

$$R = \frac{J_{ph}}{P}, \quad (2)$$

where $J_{ph}$ is the photocurrent, and P is the power of the incident light. It is observed that the responsivity of $GO/Ga_2O_3$ composite sensor increases from 20 mA/W to 90 mA/W when the applied voltage changes from 2 V to 10 V. The device fabricated in this study has acceptable performance as compared to others that do not use expensive materials [19].

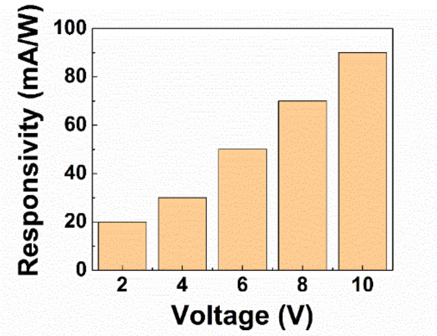


**Figure 7.** *Responsivity – voltage characteristics of $GO/Ga_2O_3$ composite sensor excited by the wavelengths of 650 nm.*

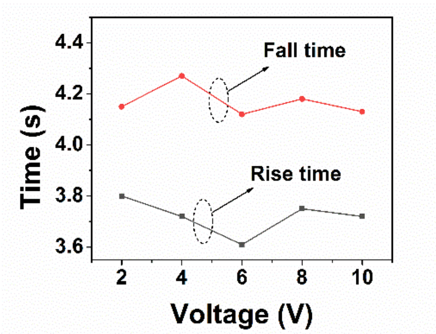


**Figure 8.** *Rise time and fall time of $GO/Ga_2O_3$ composite sensor as a function of bias voltages, excited by the wavelengths of 405 nm and 650 nm, respectively*

It was suggested that the dominant mechanism of photocurrent generation in GO-based photodetectors is controlled by the photoconductive effect [23]. The overall process is (i) carriers to be generated as $GO/Ga_2O_3$ film adsorbs the incident light, (ii) transportation of carriers and (iii) collection of carriers through the external circuit. These factors affect the rise time and the photocurrent of the optical sensors. Figure 8 shows the rise time and the fall time of the $GO/Ga_2O_3$ composite optical sensor extracted from Figure 5. The rise time ($t_1$) is calculated from 10% to 90% amplitude of the dark current, and recovery (fall) time ($t_2$) is from 90% to 10% amplitude of light current. The average rise time is ~ 3.7 s when the voltage changes from 2 V to 10 V, 10% less than the fall time.

## 4. Conclusions

Graphene oxide (GO) was exfoliated employing electrochemical method, and $Ga_2O_3$ was successfully deposited on GO via hydrothermal method. Material characterization method FESEM confirmed the two-dimension (2D) structure of GO and the formation of GO/$Ga_2O_3$ composite. The optical sensor fabricated from GO/$Ga_2O_3$ composite revealed the repeatable and reliable properties. The photocurrent was obtained to be ~ 9 µA at applied voltage of 10 V under the excitation of wavelength of 650 nm, illustrating the potential applications in optical sensing. The rise time and fall time of GO/$Ga_2O_3$ composite was detected to be ~ 3.7 s and 4.2 s. The results in this study provide a capability to use graphite rods of disposed zinc batteries to produce valuable product such as optical sensors.

## Acknowledgments

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## Conflict of Interest

The authors declare no conflict of interest.

## Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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