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# PREPARATION OF REDUCED GRAPHENE OXIDE: APPLICATION IN A PATTERNED TEMPERATURE SENSOR

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***Abstract***

***In this study, an efficient way of fabricating a reduced graphene oxide (rGO) patterned temperature sensor and its sensing properties was demonstrated. The sensor device was fabricated by drop casting the GO solution directly on the platinum interdigital electrodes (Pt-IDE) and was allowed to dry and then heated at about 400 ºC for few minutes then followed by hydrazin vapor treatment. Profilometrry, UV-VIS spectroscopy and Hall effect measurement was used to characterize the structural properties of this device. The temperature sensing properties of rGO were meassured from 35- 150 ºC and it was found that the resistance decreases exponentially with increasing temperatrure which is similar to that of a standard thermistor. The high sensivity, stability and repeatability of the rGO sensor observed are favourable for its patterned temperature sensing applications.***

***Keywords*:** Graphene Oxide, Patterned Temperature, Sensor

## 1.0 INTRODUCTION

Since the discovery of graphene, it has become an extensively research material due to its reported range of physical properties back in 2004 and 2005 [1], [2], [3], [4]. It is formed by a thick sheet of carbon atoms bonded by sp hybridization arranged in a hexagonal array [3]. It has gained interest in various fields of science and technology due to its unique electrochemical properties which are high thermal conductivity, high current, density, chemical inertness, optical transmittance and very high hydrophobicity [5], [6], [7] [4]. X-ray diffraction technique reveals the crystal structure of graphene as a closely packed honeycomb-like structure [8]. The graphene family are reduced graphene oxide (rGO), graphene oxide (GO) [9], graphene sheets and layered graphenes such as few layered graphenes and multilayered graphene (MLG) [10]. Graphene has been studied in many applications, such as electronics [11], flexible electronics [12], [13], sensors [14], [15] biomedicine [16], batteries [17], and water quality monitoring [13].

Graphene is produced in many different ways, but the two major approaches have been defined as the: top-down (TD) and bottom-up (BU) methods [18]. TD methods are generally destructive methods where a bulk (carbon) material is broken into smaller sheets of graphene [13]. The TD methods are mechanical cleavage, solution exfoliation, ball milling, electrochemical exfoliation, sonication, explosion, and shockwave exfoliation [19]. Conversely, the BU methods are constructive methods. During BU processes, graphene sheets are assembled from carbon atoms [13]. This approach is more expensive and harder to scale up, but the final graphene contains much fewer defects and is close to pristine graphene [18]. The BU methods that are used to produce graphene are epitaxial growth and chemical vapor deposition (CVD) [19], [13].

One of the many fields where graphene is studied and used in experiments is active sensor materials. The main differences in the relevant research articles are the production/preparation mechanism and graphene active layer of the sensors, which correspond to different material structures [13].

Temperature sensors play a vital role in many current and future temperature measurement applications [21], [22], [23]. Temperature sensors are manufactured to meet certain requirements. The main requirements are measurement accuracy, a good response time, and consistent performance under varying environmental conditions [13]. Many efforts has been made to develop high performance temperature sensors using carbon-based materials including carbon nanotubes, but these materials have their own advantages, shortcomings and specific working conditions [5]. Recently, graphene oxide (GO) based compounds have aroused special interest from several workers due to its various sensing applications [1]. However, the application of the conventional GO as a conductance-based sensor, makes it unfit due to the presence of various

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functional groups, whereas reduced graphene oxide (rGO) restores the conductivity by removal of these functional groups and makes it suitable for these type of sensing applications [24]. Due to its atomically thin and high surface-to-volume ratio property, its surface can absorb gas molecules very efficiently [24].
Although there have been many reports on the gas sensing properties of GO, its temperature sensing properties has not been explored much. A report has shown the infrared photodetector and wearable temperature sensor in the temperature range of 35-45 C using the reduced graphene oxide and graphene flakes [25]. A report on the temperature sensing properties of rGO over a wide temperature range of 80 K-375 K and this was demonstrated using an ice cube [7]. Yet another study showed the use of cellulose/rGO based composite films as temperature sensor in the temperature range of 25-80 C [26]. Likewise, the temperature sensing properties of rGO in the range of 100-400k was obtained using the method of drop casting of the GO solution directly on platinum inter-digital electrodes (Pt-IDE) [24].
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## 2.0 MATERIALS AND METHOD

### Materials

### Preparation/Synthesis of Graphene Oxide

The most popular method used for the synthesis of graphene oxide is the hummer's method and the modified hummer's method also known as tour's method. In the preparation of graphene oxide, the materials used are potassium per magnate which is a powerful oxidizing agent, concentrated acid- $H_2SO_4 + H_3PO_4$ and 30% $H_2O_2$
During the preparation, 360 ml of 96% $H_2O_2$ was added to 40 ml $H_3PO_4$ and cooled to a temperature > $20^0$C in an ice bath to maintain constant low reaction temperature. Therefore, 3g of graphite was added at that temperature. Then, 18g of $KMnO_4^{-8}$ was added gradually to the solution. To avoid run-away reaction, the temperature must be kept at < 40 $^0$C. Then maintain the temperature at 50 $^0$C per 12 hours.
At this step, 400 g of ice is added to the solution gotten at the clean-up step, then we then added 3 ml of Conc. $H_2O_2$.At this point the product turns bright orange. The solution was purified by washing with 0.1 M HCl. Thus, 30 ml was sonicated with graphite oxide to form graphene oxide.

### Conversion of graphene Oxide (GO) to reduced graphene Oxide (rGO)

The reduction of graphene oxide can be achieved from a number of ways - through thermal, chemical, photonic or electrochemical methods. The methods which are capable of producing the highest quality graphene tend to be more complex.
The methods adopted in this study for converting graphene oxide to reduced graphene oxide are:
1. The graphene oxide is undergo thermal reduction to a temperature > 200 ºC
2. Chemical reduction of graphene oxide with hydrazine hydrates at 100°C for 24 hours.
3. Exposing the graphene oxide to a powerful pulsed light from xenon flashtubes
4. direct heating of graphene oxide in a furnace to very high temperatures

Using chemical reduction to reduce GO is a scalable technique; however, the rGO produced by this method often has substandard electrical conductivity and surface area.

### Heating Sensors

The sensor device was fabricated by drop casting the GO solution directly on the platinum inter-digital electrodes (Pt-IDE) and was allowed to dry and then heated at about 400 ºC for few minutes then followed by hydrazine vapor treatment. It may be noted that the resistance of the GO before annealing was about few mega-ohms but changes to several ohms after reduction. The platinum electrodes were made out of Pt metal on a thin (1 mm) $Al_2O_3$ substrate using electron beam lithography. A thinner substrate was used to ensure quick thermal equilibrium between the sensor and the environment. The temperature dependent resistance was measured using a Keithley 2400 SMU meter. The sensing properties of rGO were measured using a Keithley 2400 SMU meter.

### Characterization of GO and rGO

The thickness of the Graphene Oxide and reduced Graphene Oxide was determined using a profilometer, the UV-VIS spectra was measured using a UV spec machine. The Hall Effect measurement and I-V characteristics of the sensor was done using a Keithley 2400 source SMU meter.

### Optical Characterization.

The optical characterization was done using a UV-VIS spectroscope as shown in the Figure 1. This measurement is done to ascertain the thin layers on the substrate's and its composite characteristics of light absorption. The spectroscope, which was attached to a computer and connected to the sample stage, was used to determine the absorbance of the GO and its composites. The GO sample was positioned beneath an optical fiber cable that was connected to the spectroscope and light source. The AVA software 7.1 was able to detect the data of the sample's absorbance and reflectance. Once this has been determined, the coefficient of light absorption of the sample can be determined.

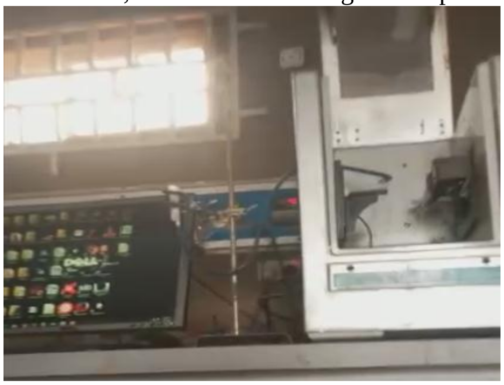

Figure1: A UV-VIS Spectroscope

**Determination of Film thickness.**

The profilometer which is a an instrument used for measuring a surface profile so as to quantify its roughness, step, curvature and flatness as shown in the Figure 2 below. The measurement can be done when the film is being deposited, in that case it is monitored or when the film has been deposited.

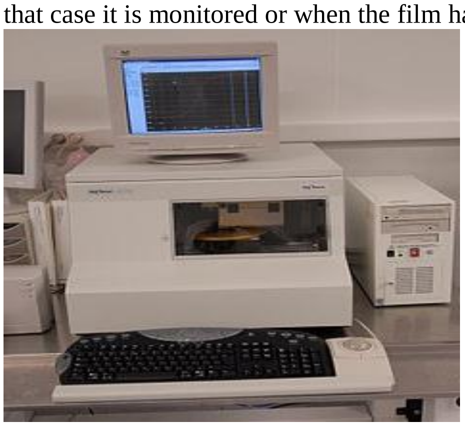

Figure 2: An image of a Profilometer Machine

**Hall Effect and I-V characterization.**

Hall Effect measurements were performed using Ecopia HMS-5000 system. The GO films were carefully deposited by a simple drop casting method so as to obtain a smooth film. For the heat sensor characterization, the I-V characteristics and the temperature dependent resistance was measured using the Keithley 2400 source SMU meter as shown in the Figure 3.


Figure 3: A Keithley 2400 source SMU meter

## 3.0 RESULTS AND DISCUSSION

**Surface Thickness of GO and rGO film**

The Figure 4 to 8 shows the profiler results indicating the film thickness of GO and rGO at a constant temperature of 50 ºC. The focal location of the profiler was selected from the XY chart at the top and bottom of the film layer so as to precisely estimate the surface height and thickness. The surface thickness of GO as shown in the Figure 4 was obtained to be 2000

$\mu m$ and that of rGO was obtained to be 2000 $\mu m$ as shown in the Figure 5. But for the Figures 6 to 8 the surface thickness for RGO_HI, RGO_HI RC and RGO_HYD was obtained to 500 $\mu m$, 1000 $\mu m$ and 1000 $\mu m$ respectively. The measurement profile was Hills & Valleys. The surface profiles are almost smooth, flat but exists only a few measurement of fluctuations. These fluctuations occurred may be due to the presence of dust particle

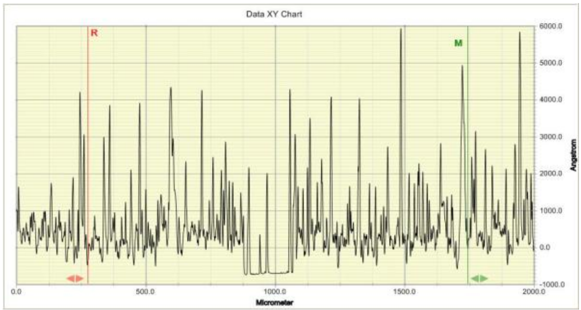


Figure 4: Surface Profiler showing the film thickness for GO

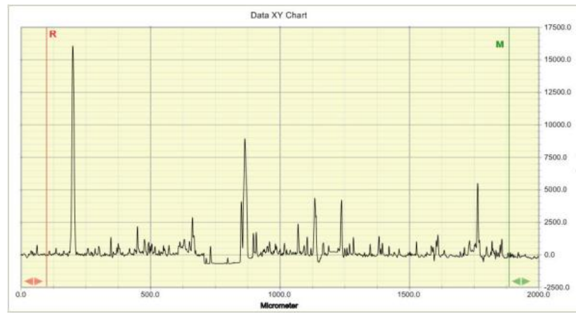


Figure 5: Surface Profiler showing the film thickness for RGO

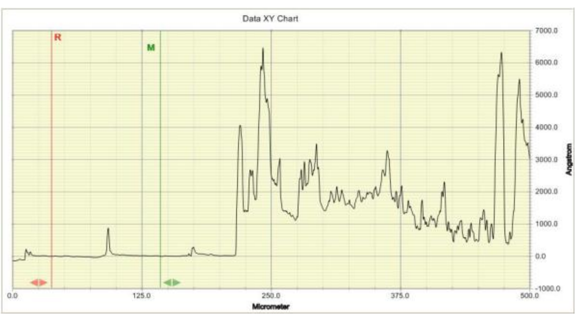


Figure 6: Surface Profiler showing the film thickness for RGO_HI

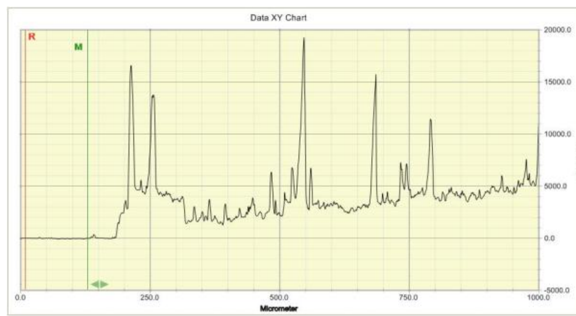


Figure 7: Surface Profiler showing the film thickness for RGO_HI_RC

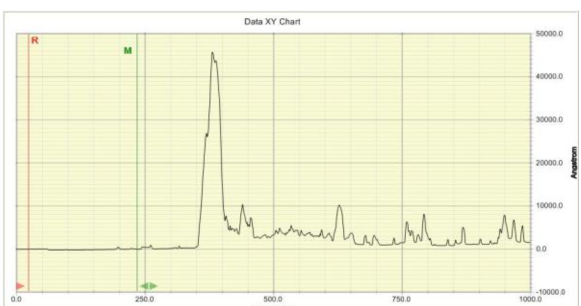


Figure 8: Surface Profiler showing the film thickness for RGO_HYD

**Temperature -resistance measurement of Heat sensor**

Table 1 shows the temperature-resistance measurement of the heat sensor fabricated from the rGO sheet. The result showed a pattern such that as there is an increase in the temperature, there is an exponential decrease in the resistance of the sensor. Figure 10 shows a plot of the temperature against the resistance as measured from the Keithley 2400 source SMU meter.

**Table 1: Temperature-Resistance relationship of the Heat Sensor**

| Temperature ( $^{0}$C) | Resistance ( $\Omega$ ) |
|---|---|
| 35.0 | 3450.778 |
| 40.0 | 3111.503 |
| 45.0 | 2870.570 |
| 50.0 | 2660.853 |
| 55.0 | 2450.662 |
| 60.0 | 2330.700 |
| 65.0 | 2190.231 |
| 70.0 | 2080.565 |
| 75.0 | 1987.592 |
| 80.0 | 1913.140 |
| 85.0 | 1841.861 |
| 90.0 | 1772.979 |
| 95.5 | 1713.486 |
| 100.0 | 1648.320 |
| 105.0 | 1595.629 |
| 110.0 | 1551.314 |
| 115.0 | 1499.067 |
| 120.0 | 1452.706 |
| 125.0 | 1431.639 |
| 130.0 | 1379.943 |
| 135.0 | 1333.128 |
| 140.0 | 1351.171 |
| 145.0 | 1301.419 |
| 150.0 | 1267.798 |

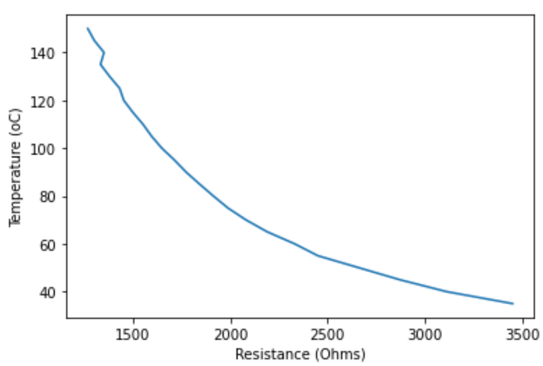


Figure 9: T-R characteristics of the Heat Sensor

As shown above, the R-T curve showed the sensitivity of the sensor. It showed an exponential decrease in the resistance as there is an increase in the temperature but this decrease is seen to occur slowly. This behavior is patterned as it is similar to that of a standard thermistor. This simply shows that the rGO based sensor exhibit an excellent sensing property for cooling and warming at different temperatures, establishing it as a potential candidate for pattern temperature sensing application.

## 4.0 CONCLUSION

In this study, the reduced graphene oxide has been prepared and thus the graphene oxide and reduced graphene oxide has been characterized to find their standard properties for application to temperature sensing. The patterned temperature sensing properties of rGO based sensor has been studied. The resistance of the rGO senor decrease exponentially with increasing temperature, which is similar to that of a standard thermostat. This sensor in this study has been established to be a potential candidate for temperature sensing applications as it has been found to exhibit an excellent sensing property for cooling and warming at different temperature.

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