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STUDY OF ELECTRICAL PROPERTIES IN GRAPHENE OXIDE THINFILMS AS A TRANSPARENT CONDUCTIVE MATERIALS

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Abstract
Graphene oxide films were successfully prepared via the modified Hummer method. The deposition of the material was done by the spin coating method. The optical characterization (Ultraviolent (UV) spectroscopy) was carried out using a (UV 752) ultraviolet-visible-Near Infra-Red (UV-VIS-NI) spectrophotometer, at the wavelength interval of 250 nm to 1100 nm. The carrier density, carrier concentration, electrical conductivity, resistivity, sheet resistance, and mobility of graphene oxide as a transparent conducting material were investigated using the Hall Effect measurement. The results are shown in a table in this work. Profilometry results showed the thickness of these reduced graphene composite films ranging from 130 – 900nm, which shows that the more layers, the better transparency it produces.

Keywords: Composite rGO Thin Films, Transparent Conducting material, Sheet Resistance absorption coefficient, refractive index, electrical properties.

Introduction

Semiconductors serve as the fundamental building blocks of contemporary electrical devices. The field of quantum physics is closely connected to the study of semiconductors, as it elucidates the behaviour of electrically charged particles as they traverse materials with a crystalline lattice structure. This relationship allows for a comprehensive comprehension of the distinctive characteristics exhibited by semiconductors. Copper and graphite, which are both considered semiconductor materials, can facilitate the movement of electrical charges between conductors and insulators such as wood and plastic (Bedi & Singh, 1998). Semiconductor materials generally exhibit greater temperatures compared to metals as a result of their enhanced electrical conductivity. Semiconductor devices possess essential characteristics that enable the passage of current in a unidirectional manner, hence demonstrating their sensitivity to variations in temperature, light intensity, resistance, and voltage. The chemical reduction of graphene oxide is considered to be a highly dependable method for the large-scale synthesis of graphene.

Graphene oxide (GO), also known as Graphitic acid or oxide, is a carbon-based chemical that consists of carbon, oxygen, and hydrogen in different proportions. It is typically produced through the oxidation of graphite using potent oxidising agents. The process of oxidising graphite results in the introduction of oxygenated functionalities into the graphite structure. This leads to the material being hydrophilic and also causes an expansion in the spacing between layers (McAllister et al., 2007). The utilisation of sonication in an aqueous medium enables the exfoliation of graphite oxide, leading to the formation of either individual or a small number of layers, commonly referred to as graphene oxide (GO). The graphite layer exhibits a complex multilayer structure, whereas graphene oxide is characterised by the presence of only a limited number of flakes, often consisting of a single layer. Graphene oxide is commonly characterised as an electrical insulator of its electrical conductivity, primarily attributed to the perturbation of its sp2 bonding network. To effectively harness the beneficial properties of its crystalline lattice structure and its electrical conductivity, it is imperative to achieve a successful reduction of graphene oxide to graphene. Once the oxygen has been effectively eliminated by the procedure, the attainment of a dispersed reduced graphene oxide (rGO) becomes more challenging due to its propensity to generate further clusters.

The graphene obtained from the reduction of graphene oxide has inherent structural and chemical vulnerabilities, which pose challenges for certain applications. However, these weaknesses also confer advantages for various other potential uses.

The characteristics of graphene oxide exhibit significant variations based on the level of oxidation and the technique of manufacture. The manipulation of graphene oxide's electrical and optical properties can be effectively achieved by regulating the concentration of oxygen-containing functional groups by various chemical reduction techniques or physical reduction approaches (Chen et al., 2012). Graphene oxide exhibits significant promise as an emerging class of nanomaterials for the production of versatile composite or synthesised materials in the form of thin films or paper-like structures. Both graphene oxide and reduced graphene oxide have emerged as significant derivatives of graphene in terms of their value and potential applications. Nevertheless, these two entities exhibit significant disparities in terms of their structural and chemical characteristics. As previously indicated, a key distinction between GO and rGO lies in the carbon-to-oxygen (C/O) ratio present inside their respective structures. The C/O ratio exhibits a notable decrease in GO structures, whereas it demonstrates a substantial increase in rGO structures, ultimately reaching an oxygen content close to zero (Ling et al., 2019). The remaining distinctions between GO and rGO materials primarily arise from variations in their carbon-to-oxygen (C/O) ratios. The primary distinction is widely regarded as the disparity in electrical conductivity between these two materials. Graphene oxide (GO) exhibits insulating or semi-conducting properties, but reduced graphene oxide (rGO) demonstrates a significantly higher electrical conductivity of 6300 S cm−1. According to the study conducted by Suresh and Solay in 2016, the disparity in the conductivities of graphene oxide (GO) and reduced graphene oxide (rGO) presents distinct opportunities for the use of these materials in various applications. One notable distinction between GO and rGO structures is their respective particular surface areas. GO exhibits a comparatively reduced surface area of 890 m2g-1, but the rGO structure nearly reinstates the exceptionally high surface area observed in pristine graphene, around 2600 m2 g−1. Singh et al. **(2**016) have reported that the mechanical strength of graphene oxide (GO) is comparatively lower than that of reduced graphene oxide (rGO). The distinct characteristics of graphene oxide (GO) and reduced graphene oxide (rGO) have been widely utilised in both academic research and industrial sectors for various purposes. (McCoy et al., 2019).

Material and Methods

The materials used are graphite powder, sodium nitrate, sulphuric acid, kmno4, thermometer and profilometer.

Synthesis of Graphene Oxide

The precursor for graphene preparation involved the synthesis of Graphene Oxide (GO) from graphite powder using a modified version of Hummer's process (Shahriary & Athawale, 2014). In summary, a mixture was prepared by combining 3g of graphite with 0.5g of sodium nitrate. A volume of 23 ml of concentrated sulfuric acid was introduced into the mixture while maintaining continuous agitation. After 60 minutes, in an environment with a temperature below 30°C, we incrementally introduced 18 grammes of KMnO4 to the pre-existing amalgamated solution as a precautionary measure against excessive heat generation and potential detonation. The combination was subjected to stirring at a temperature of 50°C for 12 hours. Subsequently, the resulting solution was diluted by the addition of 400ml of water in the form of ice, with vigorous stirring. To facilitate the full progression of the reaction involving KMnO4, an additional treatment was administered using a 3ml solution of H2O2. The resultant mixture was subjected to a washing process using 200 mL of hydrochloric acid (HCl) and 200 mL of water (H2O). Following a period of 5 minutes, the mixture was subjected to a washing process utilising 200ml of ethanol. Subsequently, the solution was subjected to filtration and subsequent drying. At this juncture, the graphene oxide was acquired. To obtain graphene, the solution was allowed to undergo a settling process for 24 hours. The removed portion, which is the graphene, exhibits a black appearance.

Growing Thin Film of Graphene Oxide

A total of five samples of the masked glass substrate were made, and a solvent was created by combining seven millilitres of Acetone with three millilitres of water. Subsequently, a homogeneous mixture was prepared by combining 2 ml of graphene with 1 ml of the solvent, ensuring thorough dispersion of both constituents. The dispersed mixture was applied onto each glass substrate sample using the spin coating method, followed by a 30-second spin-drying process using a centrifuge machine. One of the benefits associated with spin coating is the ability

to achieve consistent and uniform deposition of graphene oxide onto films. To ensure the deposition of several layers, we implemented a thorough spin-drying process for each application of graphene oxide. Additionally, we subjected the samples to annealing for 15 minutes following the last application of graphene oxide on each substrate. The black mask tape was removed before the annealing process. To determine the thickness of the graphene oxide thin film on each of the 5 glass substrates, we applied 1, 3, 5, 7, and 9 drops of graphene oxide onto each substrate.

Number of layers of 5 samples
G7 (Sample 1) -------1 layer
G 13 (Sample 2) -----3 layers
G 12 (Sample 3) -----5 layers
G 3 (Sample 4) -------7 layers
G 15 (Sample 5) -----9 layers

Annealing the Films (thermal reduction)
To become reduced graphene oxide, it must undergo either of the following reduction processes:
a. Chemical reduction process
b. Thermal/Heat reduction process
c. Photochemical reduction process

Annealing is a thermal treatment technique employed in metallurgy, wherein a metal or alloy is subjected to controlled heating at a specific temperature for a designated duration, followed by a gradual cooling process, often facilitated by furnace cooling. In this study, the films underwent an annealing process and showed the ability to withstand temperatures ranging from 250°C to 400°C for 30 minutes each. There is a positive correlation between temperature and the conductivity of the films, indicating that higher temperatures result in improved conductivity.

Characterization Techniques
Profilometry Measurement: The measurement of the profilometry, namely the thickness, of the decreased graphene thin films was performed utilising a profilometer known as the Dektak 150, manufactured by Veco instrument TMCU.S. The present study involved the implementation of profilometry testing to ascertain the optimal number of graphene deposition layers for achieving desired outcomes. The findings indicated the correlation between the thickness of each film and its subsequent influence on the performance of the film.

Optical Characterization (UV-VIS Spectroscopy): The optical analysis of the films was conducted using a UV-VIS-NI spectrophotometer (model UV 752) in the United Kingdom. The wavelength range examined was from 230 nm to 1100 nm. Ultraviolet spectrophotometry was employed as the fundamental technique for this study. Spectrophotometry is a technique that involves the absorption of light within the immediately adjacent visible range. The spectrophotometer was utilised to acquire absorbance data, while other optical parameters such as transmittance, reflectance, refractive index, absorption coefficient, extinction coefficient, and energy band gap were assessed through the application of the following equations:
The transmittance (T) of the cells was evaluated using Equation (2.1) which according to Ling (2019) is given by.

$$T = 10^{-A} \quad (2.1)$$

Reflectance was obtained using equation (2.2) which is given by

$$R = 1 - (A + T) \text{ (Ling, 2019)} \quad (2.2)$$

The refractive index of the cells was calculated using Equation (2.3) as given by

$$\eta = \frac{(1+\sqrt{R})}{(1-\sqrt{R})} \text{ (Chen et al., 2012) \& (Ilenikhena, 2008)} \quad (2.3)$$

The absorption coefficient was calculated from absorbance spectra using Equation (2.4) Where *d* is the thickness and is measured in micrometres (μm), and *A* is the absorbance.

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$$\alpha = \frac{2.303A}{d}$$ (McAllister et al., 2007) (2.4)

The extinction coefficient was obtained using Equation (2.5).

$$K = \frac{\alpha\lambda}{4\pi}$$ (McCoy et al., 2019) (2.5)

Optical conductivity was estimated using Equation (2.6) as given by

$$\sigma_o = \frac{\alpha\eta c}{4\pi}$$ (Ilenikhena, 2008) (2.6)

Where c is the speed of light

The energy band gap was estimated using Tauc's model given in Equation (2.7) as given by

$$(\alpha h\nu)^n = \beta(h\nu - E_g)$$ (McCoy et al., 2019) (2.7)

Where α is the absorption coefficient, *h* is the Planck constant, v is the photon's frequency, E_g is the band gap energy and β is a constant. The *n* factor depends on the nature of the electron transition and is equal to ½ or 2 for the direct and indirect transition band gaps respectively.

Hall Effect Measurement Test

The Hall Effect measurement equipment HMS-3000, Ecopia, was employed to assess the electrical properties of the reduced graphene oxide sheets. Hall Effect measurement devices are considered superior and more desirable compared to other testing methods because of their immunity to dirt and dust, resulting in enhanced accuracy and reliability of the obtained data. The experimental findings revealed many properties of the films, including sheet resistance (Rs), resistivity, conductivity, bulk concentration, and mobility.

Results

Measurement of the Thickness of Graphene Oxide Films

The thickness of the reduced graphene oxide (rGO) films was determined using a profilometer. The thickness of sample G7 was found to be 1,300Angstrom and was converted to a nanometer (130nm), sample G12 was found to be 3000Angstrom and was converted to nanometer (300nm), and sample G15 was found to be 6000Angstrom and was converted to nanometer (600nm), sample G13 was found to be 9000Angstrom and was converted to nanometer (900nm), sample G3 was found to be 4000Angstrom and was converted to nanometer (400nm), as seen in figure 1, figure 2, figure 3, figure 4, and figure 5 below. The thickness of the samples ranges from 130-900nm, which shows that the more layers, the better transparency it produces.

Optical characterization

The analysis of UV-VIS of the reduced graphene oxide (rGO) films was determined using a (UV 752) ultraviolet-visible-Near Infra-Red (UV-VIS-NI) spectrophotometer U.K with wavelength interval of 230 nm to 1100 nm as seen in figure 6. The result indicated that the reduced graphene oxide possessed a good absorption in the visible range (380~800nm), but absorption in the ultraviolet range was also slightly decreased. The results showed good photo response of rGO sheet not only in the ultraviolent range but also in the visible range, which implied the enormous potential for the application of light as in a transparent conducting material. The direct band gap can also be derived from the results. Below are the graphical representations of some properties of the optical characterization:

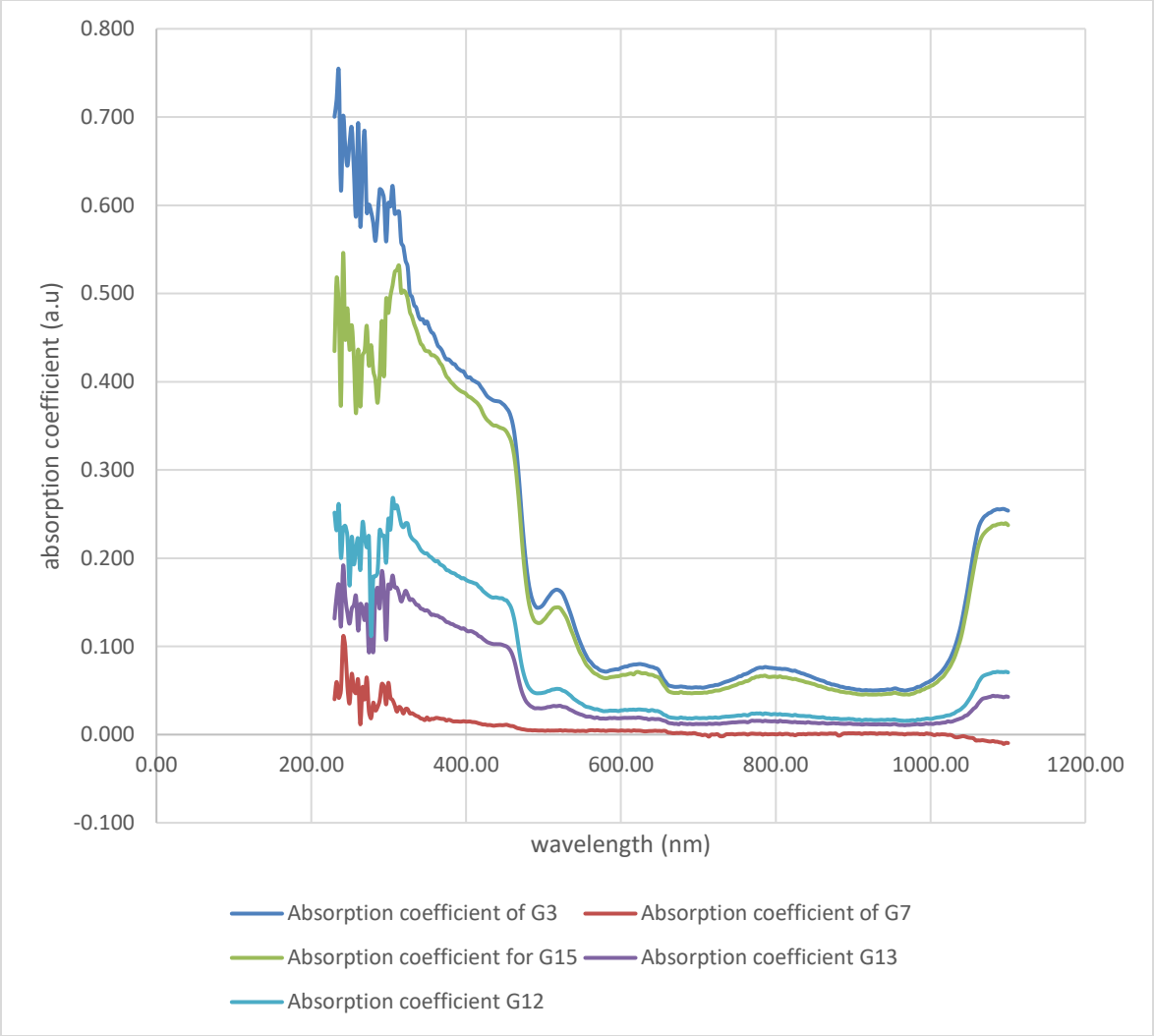

Figure 1. Absorption coefficient of the reduced graphene oxide thin films

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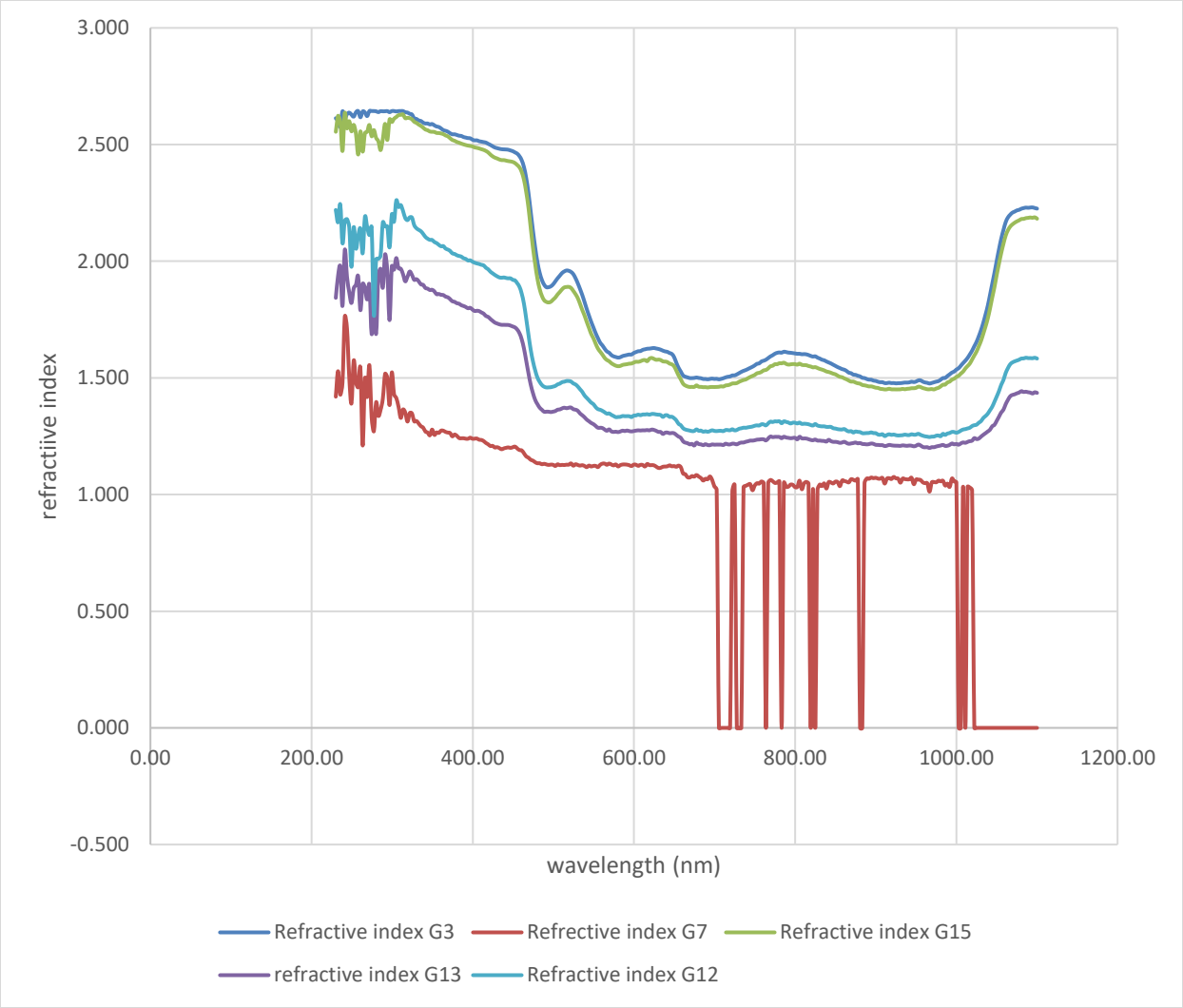

Figure 2. The refractive index of the reduced graphene oxide thin films

The absorption coefficient revealed to what extent in the layers of the reduced graphene oxide thin films, the light of a particular wavelength can enter before absorption. Fig 1. Shows that the absorption coefficient is not constant but strongly depends on wavelength, which is by results reported by Amaechi and Ogbonda (2022). The refractive index of the reduced graphene oxide thin films was investigated as seen in Figure 2. The films showed strong absorption in the visible region and poor absorption in near-infrared radiation wavelength.

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Electrical Properties: Hall Effect Results

Table 1
Hall Effect Results for the 5 samples

	Bulk concentration	**Mobility**	**Sheet Resistance**	**Resistivity**	**Sheet concentration**	**Conductivity**	**Average Hall coefficient**	**B-D cross-hall coefficient**	**The ratio of vertical/ horizontal**
	(/cm3)	(cm2/V5)	(Ω)	(Ωcm)	(/cm2)	(1/ Ωcm)	(cm3/C)	(cm3/C)	
Sample G7	-4.53E+11	2.02E+02	6.20E+09	6.82E+04	-4.98E+06	1.47E-05	-1.38E+07	-1.27E+07	1.03E-01
Sample G13	5.365+11	5.60E+01	1.04E+10	2.08E+05	1.07E+07	4.81E-06	1.16E+07	-6.23E+07	5.54E-01
Sample G12	2.48E+10	2.35E+03	3.06E+09	1.07E+05	8.68E+05	9.35E-06	2.52E+08	5.53E+07	4.48E-01
Sample G3	9.25E+14	5.42E+01	4.15E+06	1.25E+02	2.77E+10	8.03E-03	6.75E+03	1.38E+05	-1.52E+01
Sample G15	-4.01E+13	4.05E+04	8.55E+04	3.85E+00	-1.80E+09	2.60E-01	-1.56E+05	2.20E+04	8.54E-01

From the hall effect results it is observed that the rGO thin film of sample G7 (1 layer) has a very high resistivity and a low conductivity, while samples G13, G12, G3 and G15 have poor resistivity and in turn their conductivity is relatively high. It is observed that multilayers of rGO have good electrical properties.

Conclusion

The advent of Graphene has significantly influenced the scientific realm and is poised to emerge as a ubiquitous material within the electronics field shortly. To achieve effective production, it is important to generate graphene sheets on a large scale while considering crucial factors such as band gap, conductivity, resistivity, and transmittance qualities. There has been a notable increase in the utilisation of environmentally sustainable reducing agents in the synthesis of graphene-based products. The fabrication of thin films of graphene oxide has been effectively achieved by the utilisation of the modified Hummer process. Additionally, the samples must undergo an annealing step to achieve the transformation into reduced graphene oxide. The effective preparation of the films was demonstrated through manufacturing, profilometry, optical, and electrical characterisation. A systematic procedure was employed to compute the band gap, following a sequential formula.

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