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OPTICAL CHARACTERIZATION OF REDUCED GRAPHENE OXIDE FILMS AS TRANSPARENT CONDUCTIVE MATERIALS

[1]Adekanmbi, M., [2]Jonah-Iniebiy, S.I., & [3]Amaechi, C.J.
[1-3]Department of Physics, Ignatius Ajuru University of Education, Rumuolumeni, Rivers State, Nigeria.

Corresponding author email: sherpmart@yahoo.com

Abstract
Graphene oxide films were successfully prepared via the modified Hummer method. The deposition of the material was done by the spin coating method. 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. The optical characterization Ultraviolent (UV) spectroscopy was carried out using a UV 752 ultraviolet-visible-near infrared (UV-VIS-NI) spectrophotometer, at the wavelength interval of 250 nm to 1100 nm, while the Tauc model was used to obtain the optical band gap. The optical band gap of the graphene oxide films was 2.2 eV, 2.3 eV, 2.1 eV, 2.4 eV, and 2.0 eV respectively, which showed that the reduced graphene oxide films possess excellent optical response and outstanding thermal stability.

Keywords: Composite rGO Thin Films, Transparent Conducting material, Sheet Resistance Transmittance, reflectance.

Introduction

Semiconductors serve as the fundamental building blocks of contemporary modern devices. It is imperative to comprehend the characteristics of semiconductors, which are intricately linked to the principles of quantum physics. Quantum physics elucidates the movement of electrically charged particles within a solid substance, wherein the atoms or molecules are organised in a minuscule structure referred to as a crystal lattice. According to Bedi and Singh (1998), semiconductor materials, including copper and graphite, can facilitate the conduction of electrical charges between conductors and insulators such as wood and plastic. Semiconductor materials exhibit elevated temperatures due to their electrical conductivity, which differs from that of metals. Semiconductor devices possess notable characteristics that enable them to conduct current in a unidirectional manner, hence demonstrating their sensitivity to thermal radiation, light, and changing resistivity. The electrical properties of a semiconductor material can be enhanced by the deliberate introduction of impurities, hence enabling control over its overall efficacy. The process of introducing impurities into a semiconductor material is commonly referred to as "doping". Therefore, materials fabricated from semiconductors are utilised in the processes of energy conversion and switching. In the realm of semiconductors, the conduction of electric current occurs through the presence of charge carriers, which encompass both freely moving electrons and holes. Doping is a technique employed to augment the quantity of charge carriers within a semiconductor material. The term "n-type" is used to describe a doped semiconductor that predominantly consists of free electrons. Semiconductor materials employed in electronic devices undergo precise doping procedures to regulate the spatial distribution and density of p- and n-type regions. The resulting p-n junctions between these regions are accountable for the desirable electronic functionality. While certain pure elements and compounds exhibit semiconductor characteristics, silicon, germanium, and gallium compounds are the most frequently utilised elements in electronic devices (Adashi, 1994). The researchers focus their investigation on silicon, a widely utilised semiconductor material that has undergone extensive characterisation techniques commonly employed in the field. The reason for this phenomenon is attributed to the affordability and extensive utilisation of silicon in the field of computers. This will additionally serve as a stimulus for researchers to persist in their investigations about the characterization of alternative semiconductor materials suitable for application in power electronics, including LED devices, photovoltaics, and other related technologies. To identify the similarities among these materials, it will be necessary to modify numerous existing characterization methods (Baladin, 2008). Conductors are characterised by their high electrical conductivity, which is attributed to the presence of abundant free electrons and holes inside their atomic structure. Conductivity, in a broad sense, refers to the ability to conduct various forms of energy, such as electricity or heat. Insulators are characterised by their inability to convey electrical current, rendering them ineffective as conductors. These materials possess the ability to disrupt and impede the passage of electric current. Insulation is commonly employed in household products and electrical circuits to provide protection and insulation. In the context of thermal insulation, its primary function is to impede the transfer of heat rather than electrical conductivity.

Materials and Methods

The materials used are graphite, sodium nitrate concentrated sulphuric acid(h_2so_4), potassium permanganate ($kmno_4$), hydrochloric acid, water, hydrogen peroxide, acetone, and Thermometer. The method used was the Spin coating method and Modified Hummer's method.

Synthesis of Graphene Oxide

The precursor for the preparation of graphene involved the synthesis of Graphene Oxide (GO) from graphite powder, with a modified version of Hummer's process (Shahriary & Athawale, 2014). In summary, a mixture was prepared by combining 3 grammes of graphite and 0.5 grammes of sodium nitrate. A volume of 23 ml of concentrated sulfuric acid was introduced into the system while maintaining continuous agitation. After 60 minutes, in an environment with a temperature below 30°C, we proceeded to incrementally introduce 18 grammes of KMnO4 into the pre-existing mixture to mitigate the risk of 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, while maintaining a vigorous stirring process. To facilitate the full completion 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 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 component refers to the graphene material with a black appearance.

Growing Thin Film of Graphene Oxide

A total of five samples of the masked glass substrate were made for experimentation. A solvent was created by combining 7ml of Acetone with 3ml of water. Next, a mixture of 2 ml of graphene and 1 ml of the solvent was prepared, ensuring thorough dispersion of both components. 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 a consistent and uniform deposition of graphene oxide onto films. To ensure the deposition of many layers, we implemented a thorough spin-drying process to remove any excess graphene oxide and subsequently annealed the samples for 15 minutes after the final drop was applied 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 glass substrates, we applied varying quantities of drops (1, 3, 5, 7, and 9) of graphene oxides onto the substrates across five distinct samples.

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, typically 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 conducted utilising a profilometer known as the Dektak 150, manufactured by the Veco instrument TMCU.S. The present study involved the implementation of profilometry testing to ascertain the optimal number of graphene deposition layers for effective outcomes. The findings indicated the measurement of 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 near-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) given by

$$T = 10^{-A} \quad \text{(Lokhande et al., 2002)} \qquad (2.1)$$

Reflectance was obtained using equation (2.2) given by [6].

$$R = 1-(A+T) \quad \text{(Lokhande et al., 2002)} \qquad (2.2)$$

The refractive index of the cells was calculated using Equation (2.3) as given by [5] and [7].

$$\eta = \frac{(1+\sqrt{R})}{(1-\sqrt{R})} \quad \text{(Ilenikhena, 2008) \& (Ohwofosirai et al., 2014)} \qquad (2.3)$$

The absorption coefficient was calculated from absorbance spectra using Equation (2.4) given by [9]. Where *d* is the thickness and is measured in micrometres (μm), and *A* is the absorbance.

$$\alpha = \frac{2.303A}{d} \qquad (2.4)$$

The extinction coefficient was obtained using Equation (2.5)

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

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

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

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

$$(\alpha hv)^n = \beta(hv - E_g) \quad \text{(McCoy et al., 2019)} \qquad (2.7)$$

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.

Results

Measurement of the Thickness of Graphene Oxide Films

The measurement of the thickness of the reduced graphene oxide (rGO) films was conducted utilising a profilometer. The thickness measurements of the samples G7, G12, G15, G13, and G3 were determined to be 1,300 Angstrom, 3,000 Angstrom, 6,000 Angstrom, 9,000 Angstrom, and 4,000 Angstrom, respectively. These measurements were subsequently converted to nanometers, resulting in values of 130 nm, 300 nm, 600 nm, 900 nm, and 400 nm for samples G7, G12, G15, G13, and G3, respectively. These values are depicted in Figures 1, 2, 3, 4, and 5, as shown below. The samples exhibit a variety of thicknesses spanning from 130 to 900 nanometers, indicating that increased layering results in enhanced transparency. The following figures depict the graphical representations of the profilometry results.

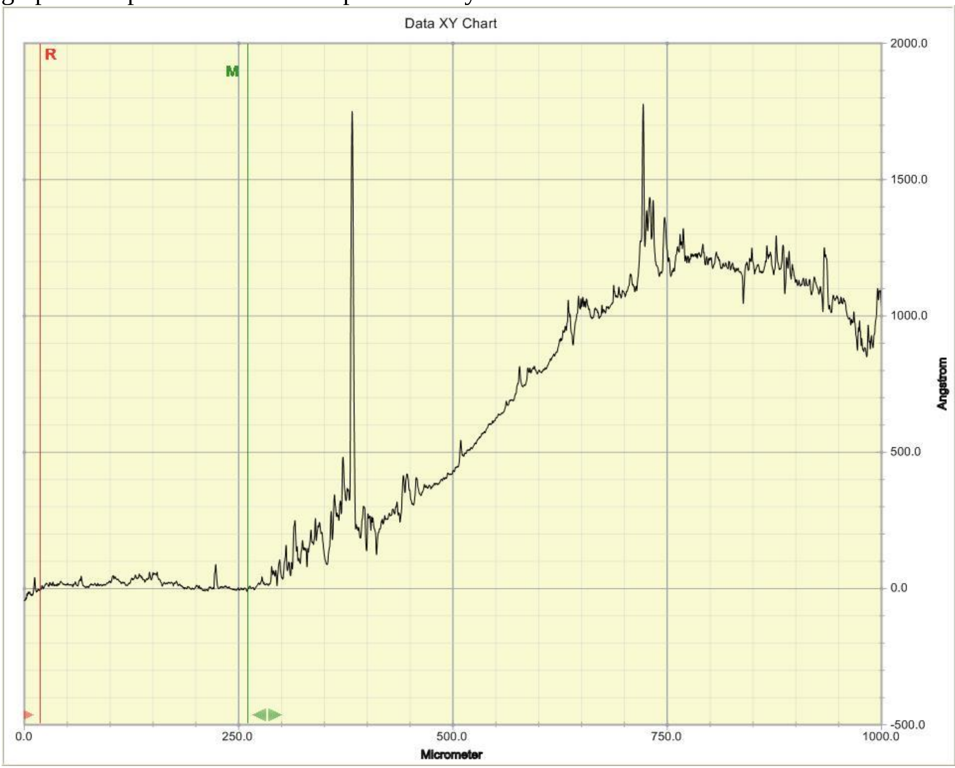

Figure 1. Thickness graph of Sample G7 1 layer

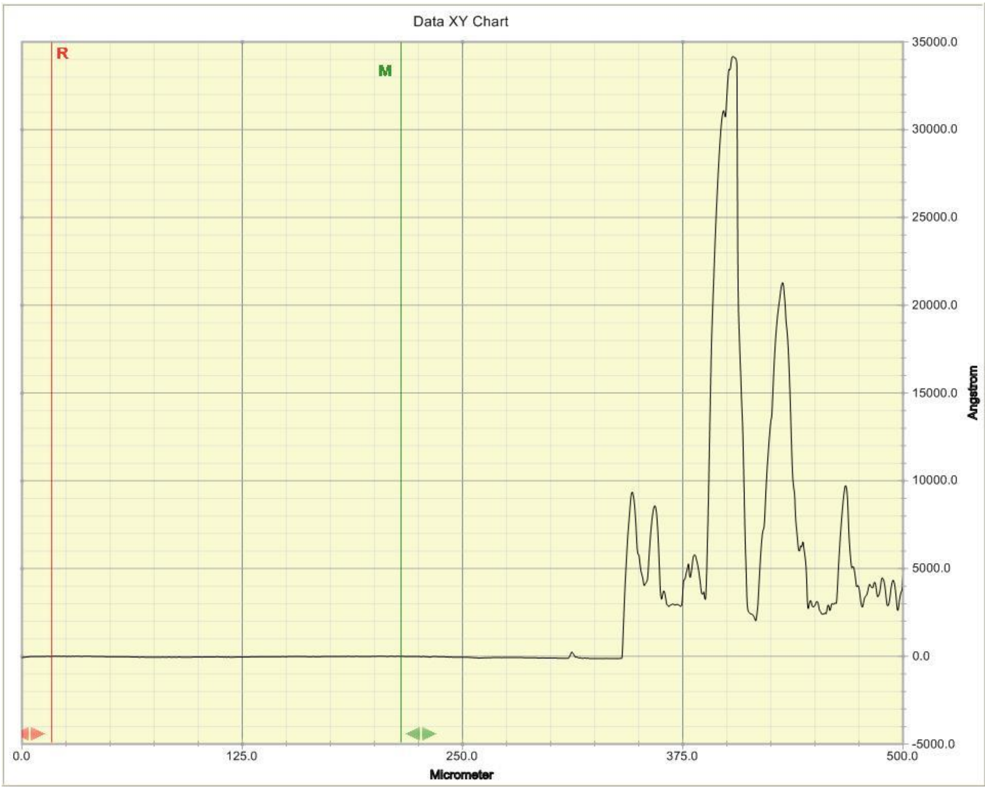

Figure 2. Thickness graph of Sample G13- 3 layers

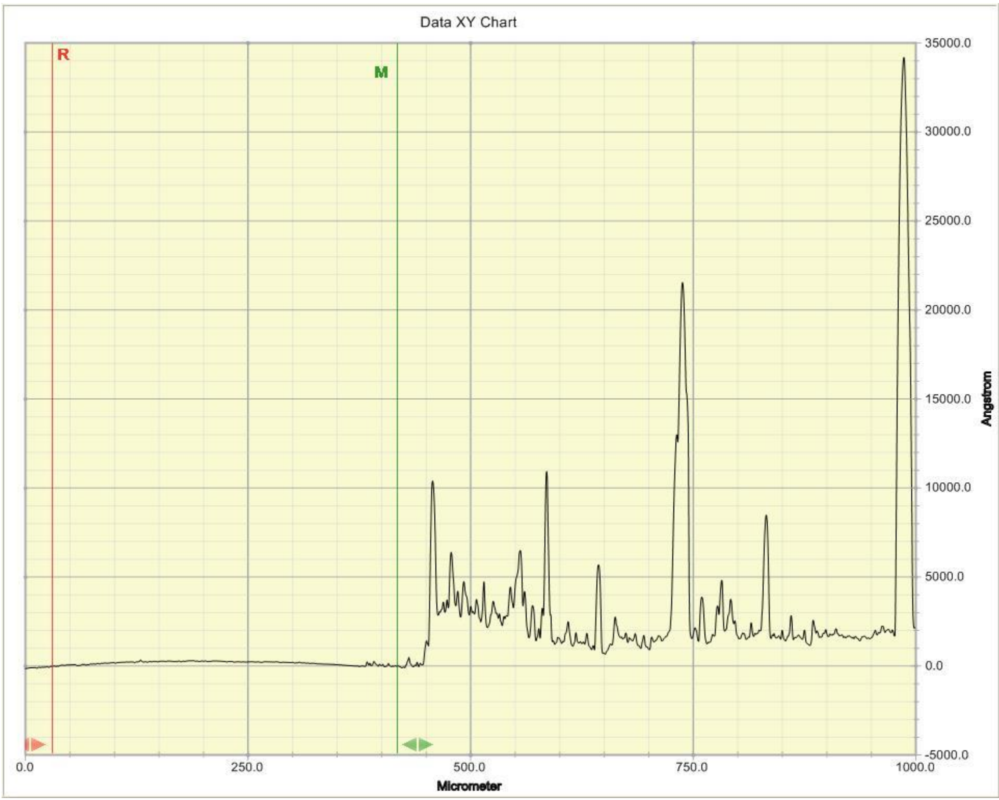

Figure 3. Thickness graph of Sample G12 - 5 layers

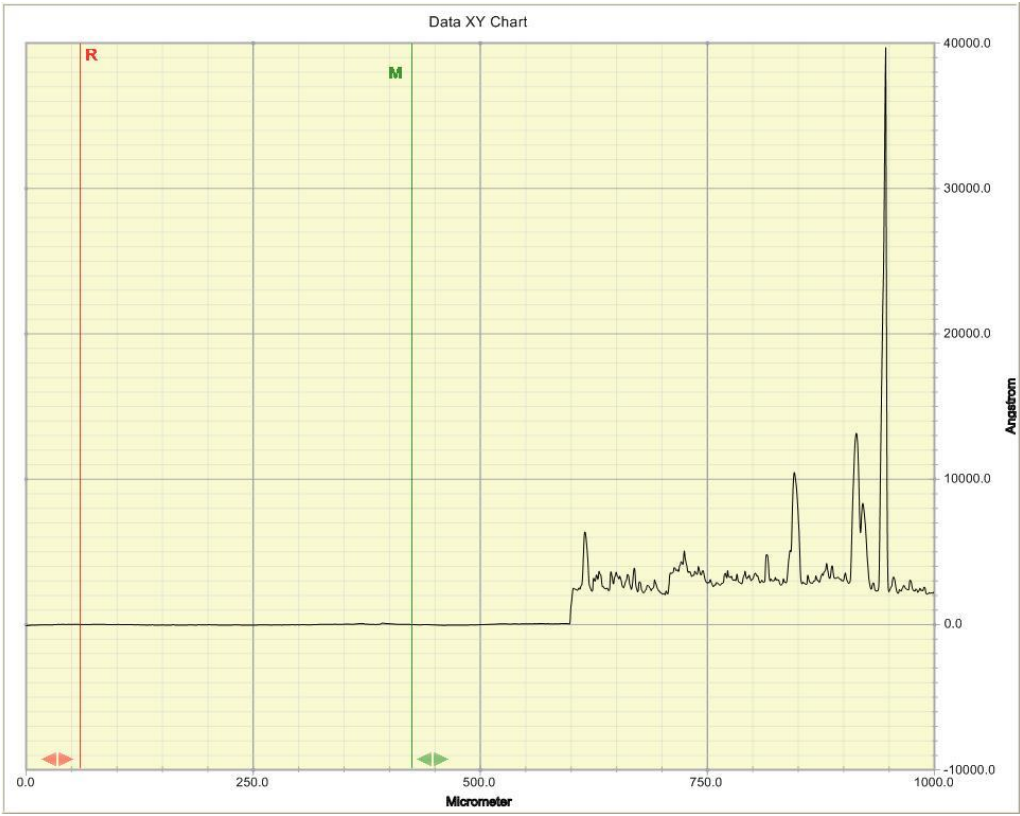

Figure 4: Thickness graph of Sample G3 – 7 layers

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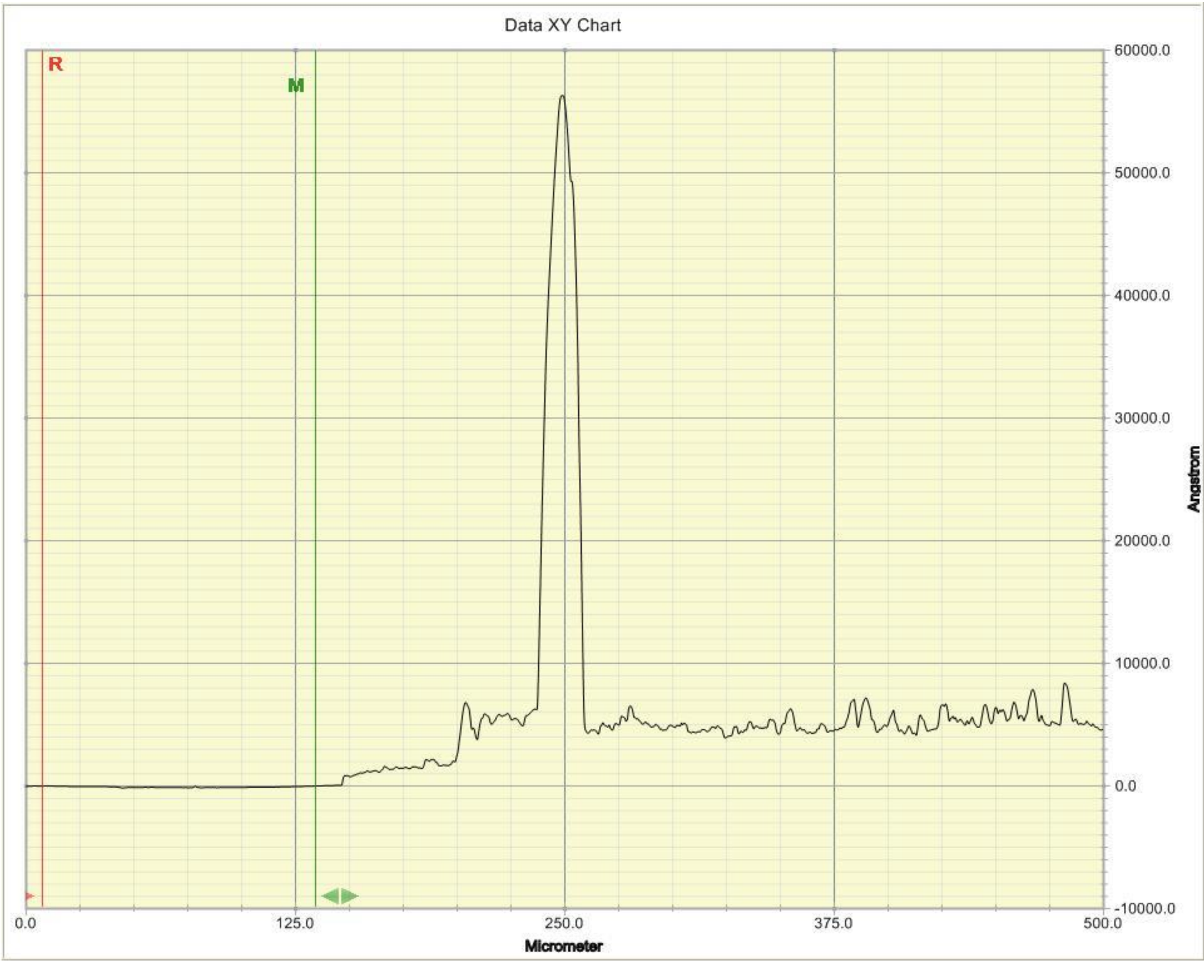

Figure 5. Thickness graph of Sample G15 - 9 layers

Optical characterization

The UV-VIS analysis of the reduced graphene oxide (rGO) sheets was conducted using a UV-VIS-NI spectrophotometer (UV 752) from the United Kingdom. The spectrophotometer has a wavelength range of 230 nm to 1100 nm, as depicted in Figure 6. The findings of the study revealed that the reduced graphene oxide had favourable absorption characteristics within the visible range (380~800nm), albeit with a minor decrease in absorption inside the ultraviolet range. The findings demonstrated that the rGO sheet exhibited a favourable photo response not only within the ultraviolet spectrum but also within the visible spectrum. This suggests significant prospects for utilising light as a transparent conducting material. The results can also be used to derive the direct band gap. The following are the visual depictions of each attribute of the optical characterisation.

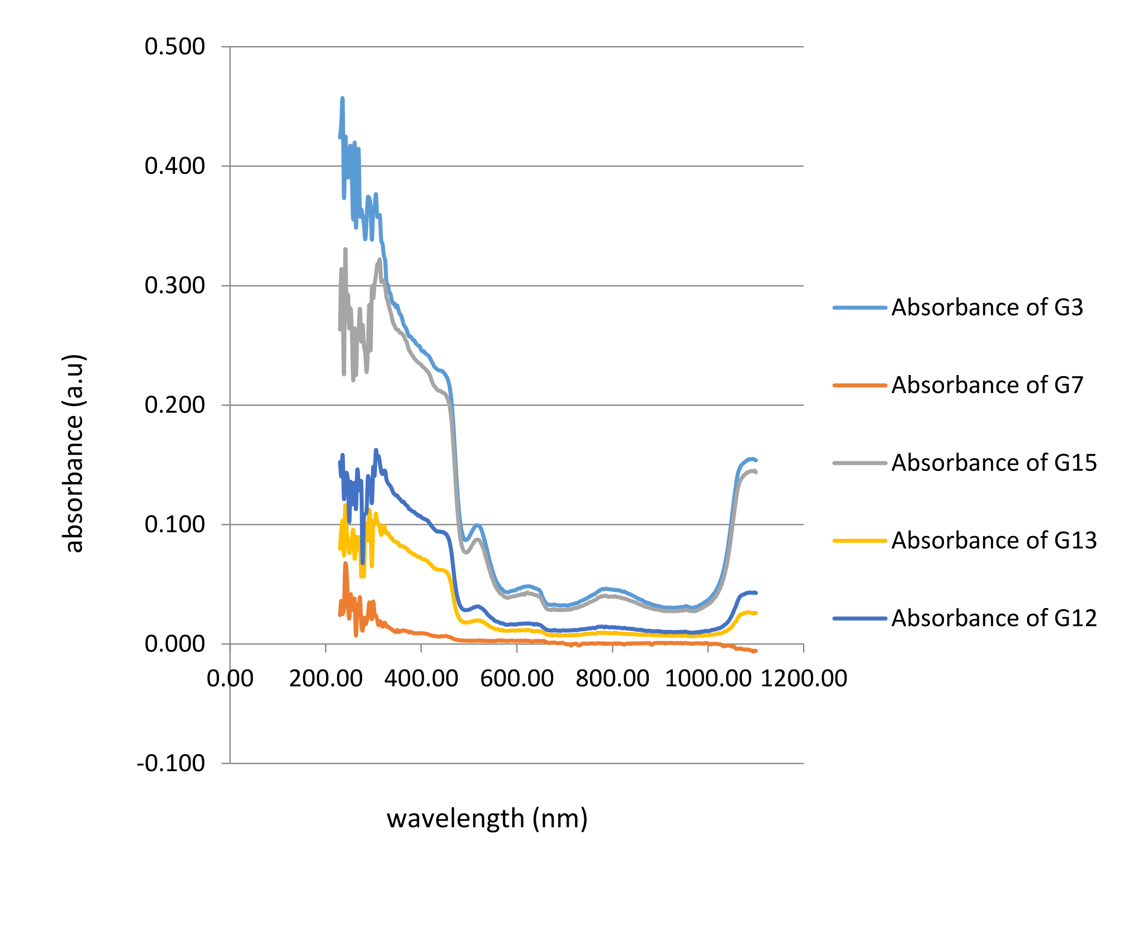

Figure 6: absorbance property of all 5 samples

The absorbance graph shows that G3 (7layers) has the best absorbance level; followed by G15 (9layers) which is almost overlapping G12, and G7 and G13 possess poor absorbance properties.

The higher the absorbance level, the better and more effective the film becomes as it can absorb the entire light incident on the film.

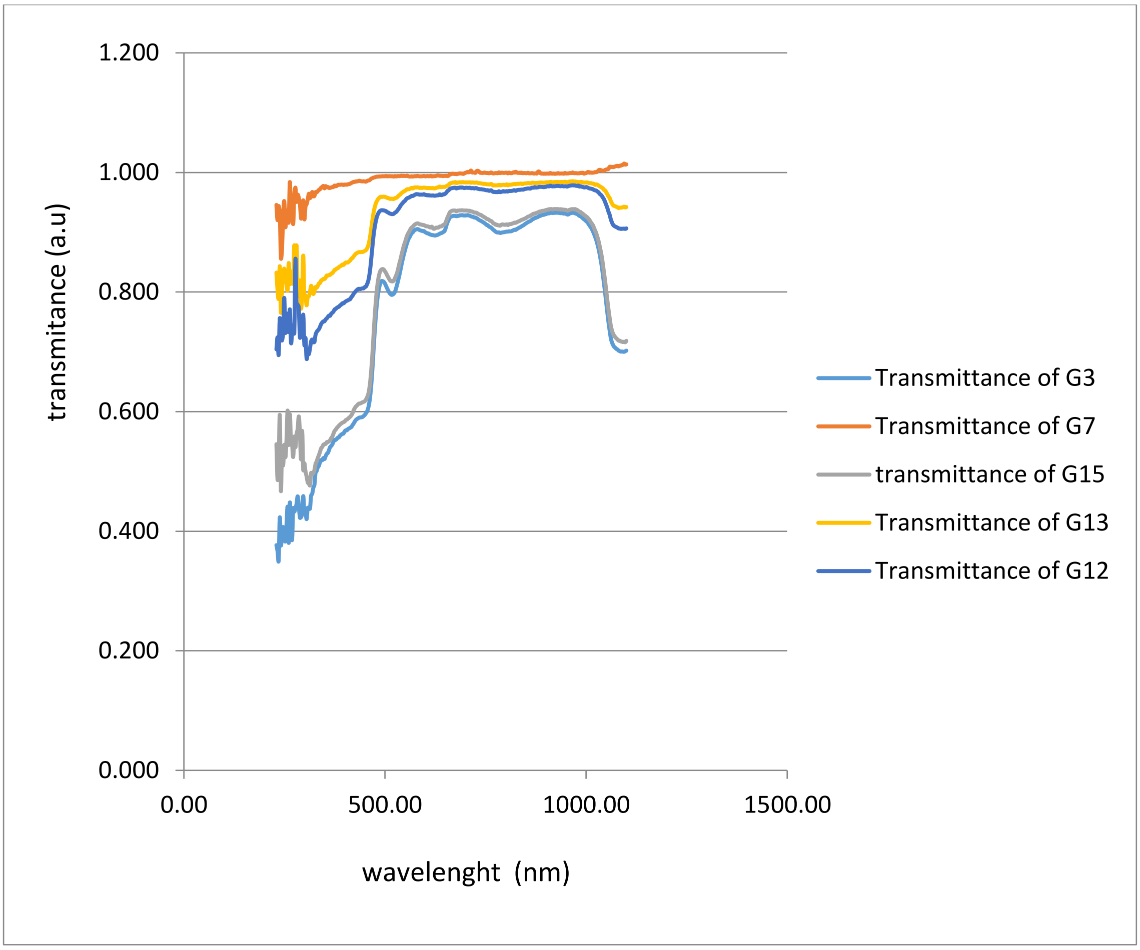

Figure 7: transmittance property of all 5 samples

The transmittance graph shows that G3 (7layers) has the best transmittance level; followed by G15 (9layers) which is almost overlapping G3, then G12 (5 layers), G13 (3 layers) andG7 (1 layer) possess poor transmittance property. The lower the transmittance, the more effective the film becomes as no light rays can pass through the film.

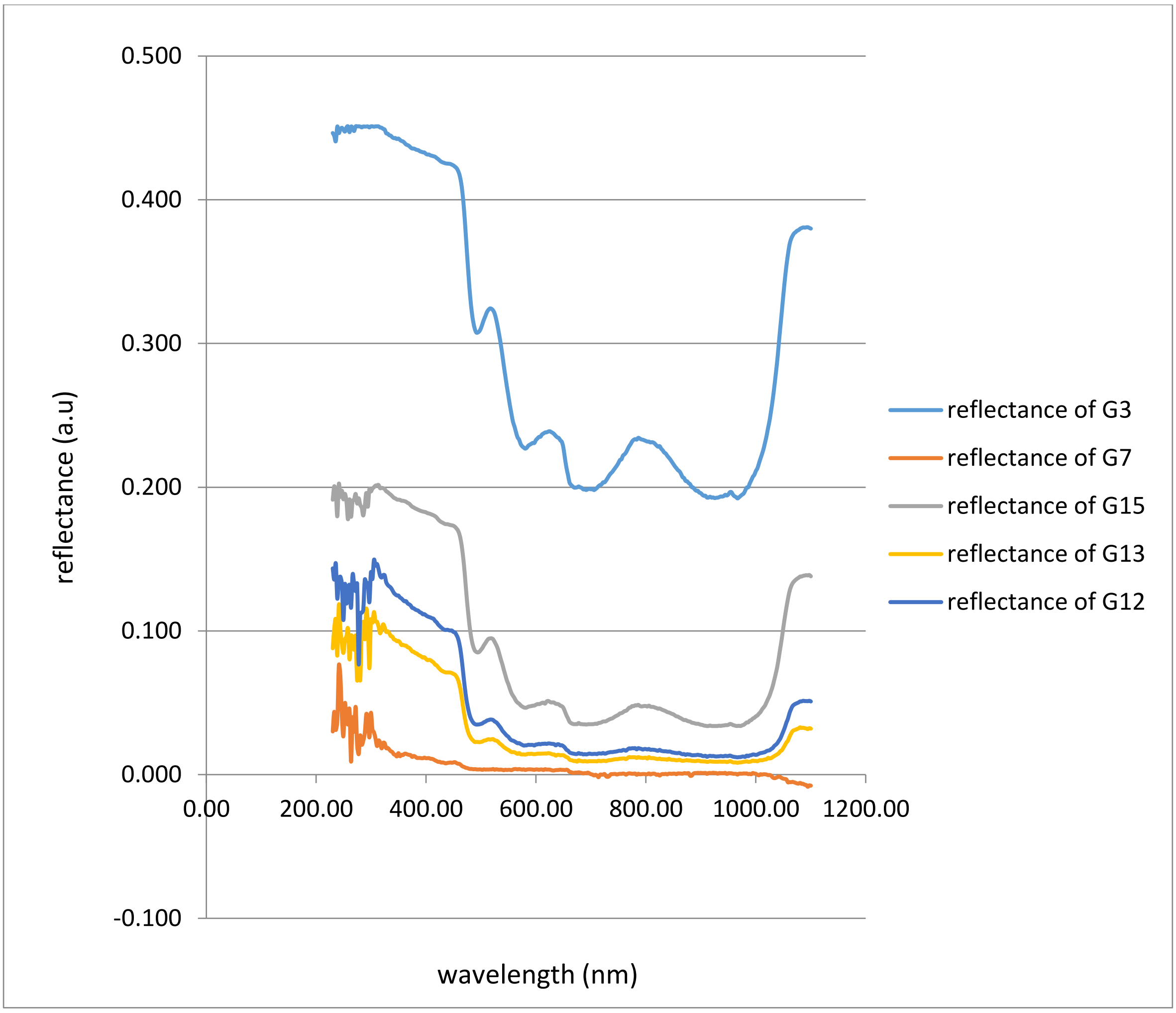

Figure 8: reflectance property of all 5 samples

The reflectance graph shows that G3 (7layers) has the best reflectance level; followed by G15 (9layers) and then G12 (5 layers), G13 (3 layers) andG7 (1 layer) possess poor transmittance property accordingly

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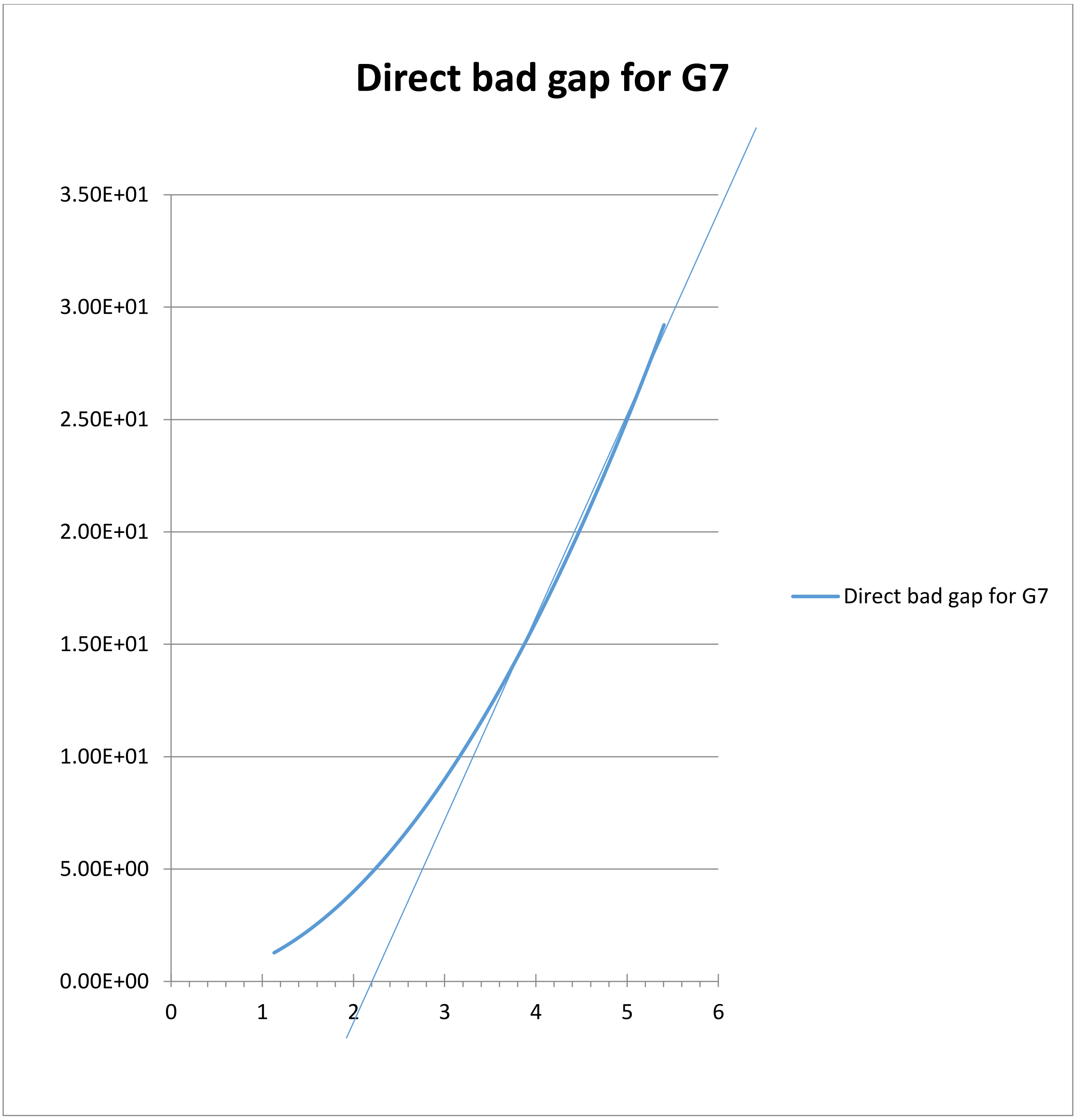

Figure 9. Band gap for sample G7 (1 layer)

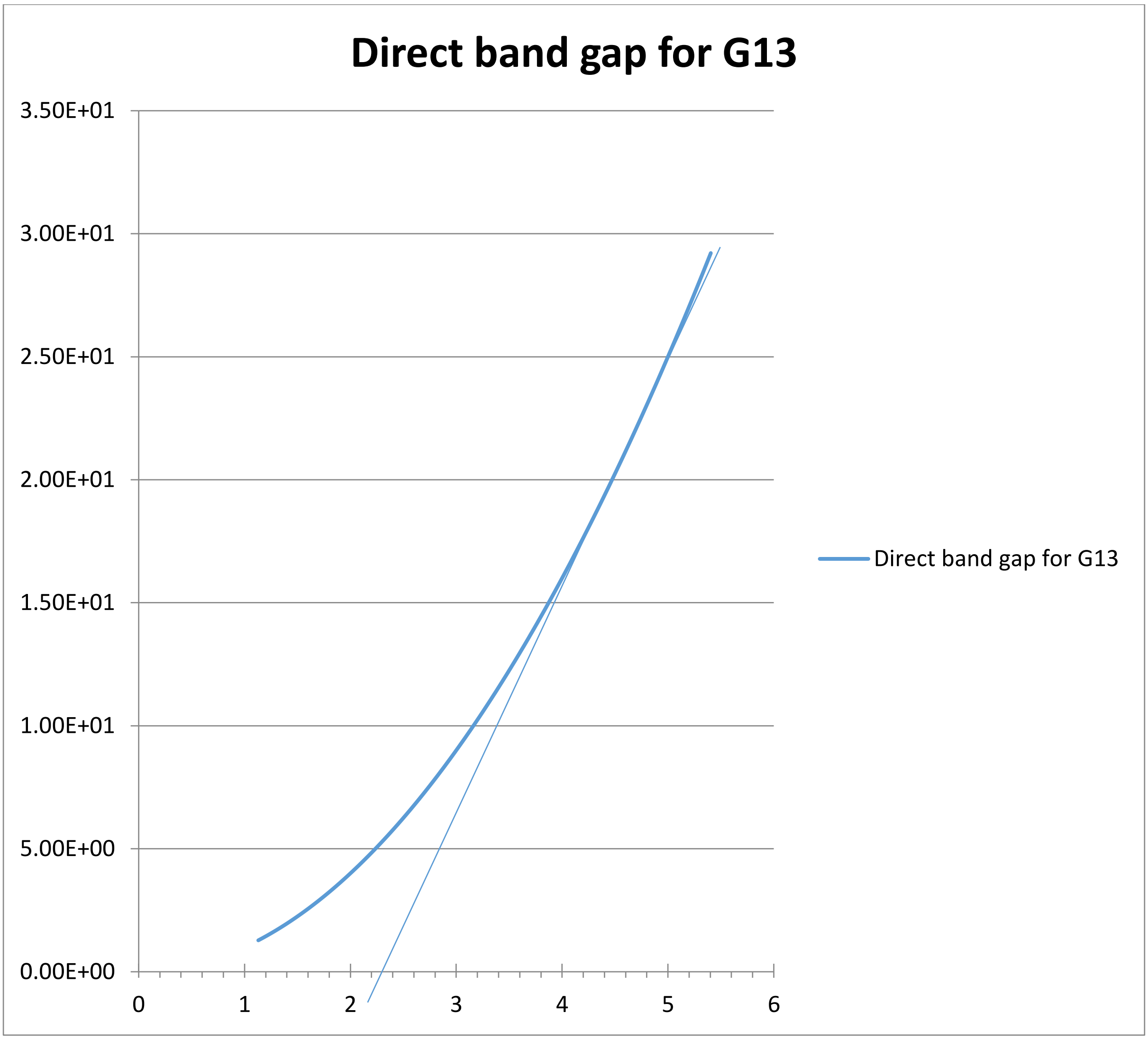

Figure 10. bandgap for sample G13 (3 layers)

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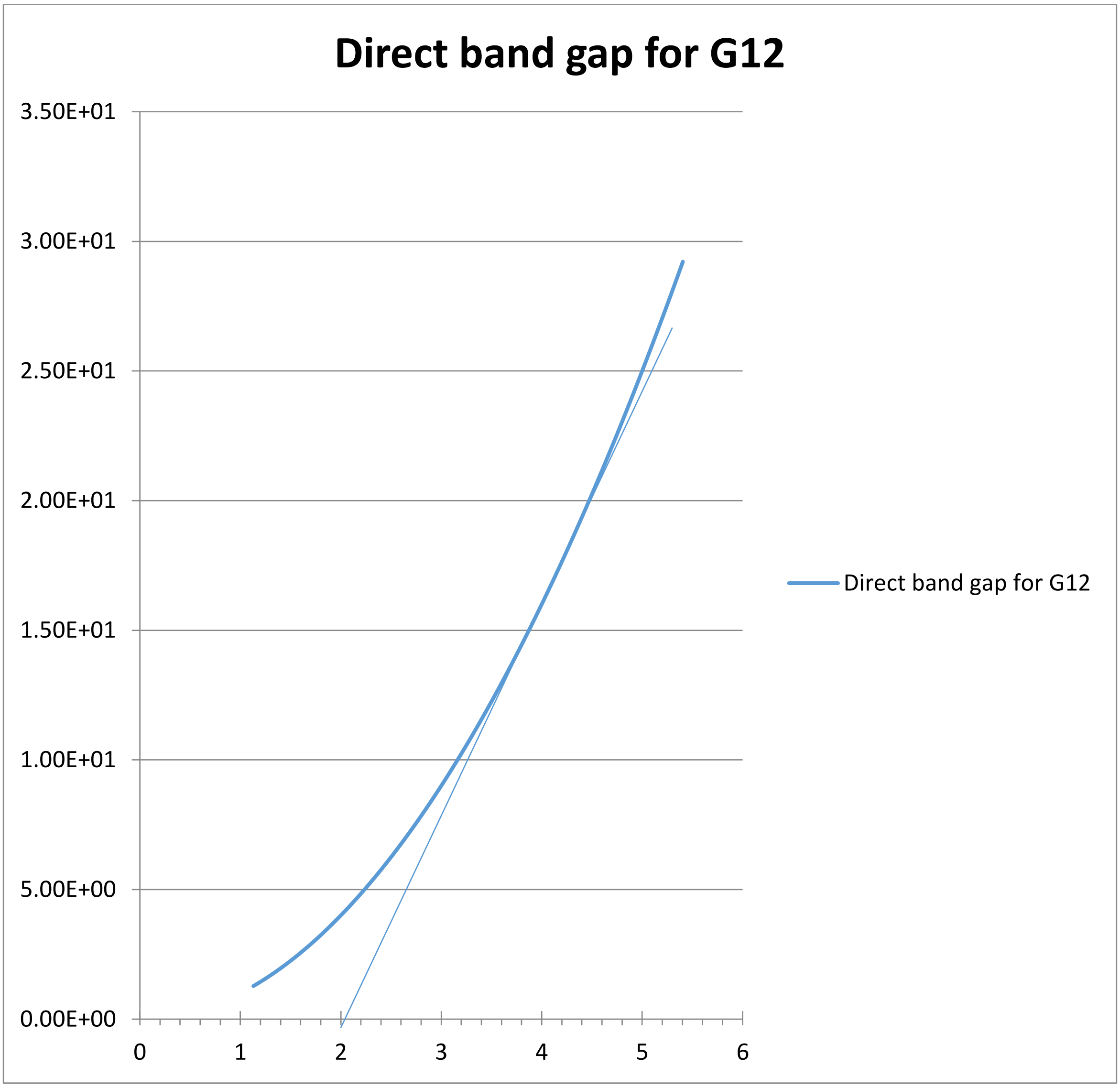

Figure 11. bandgap for sample G12 (5 layers)

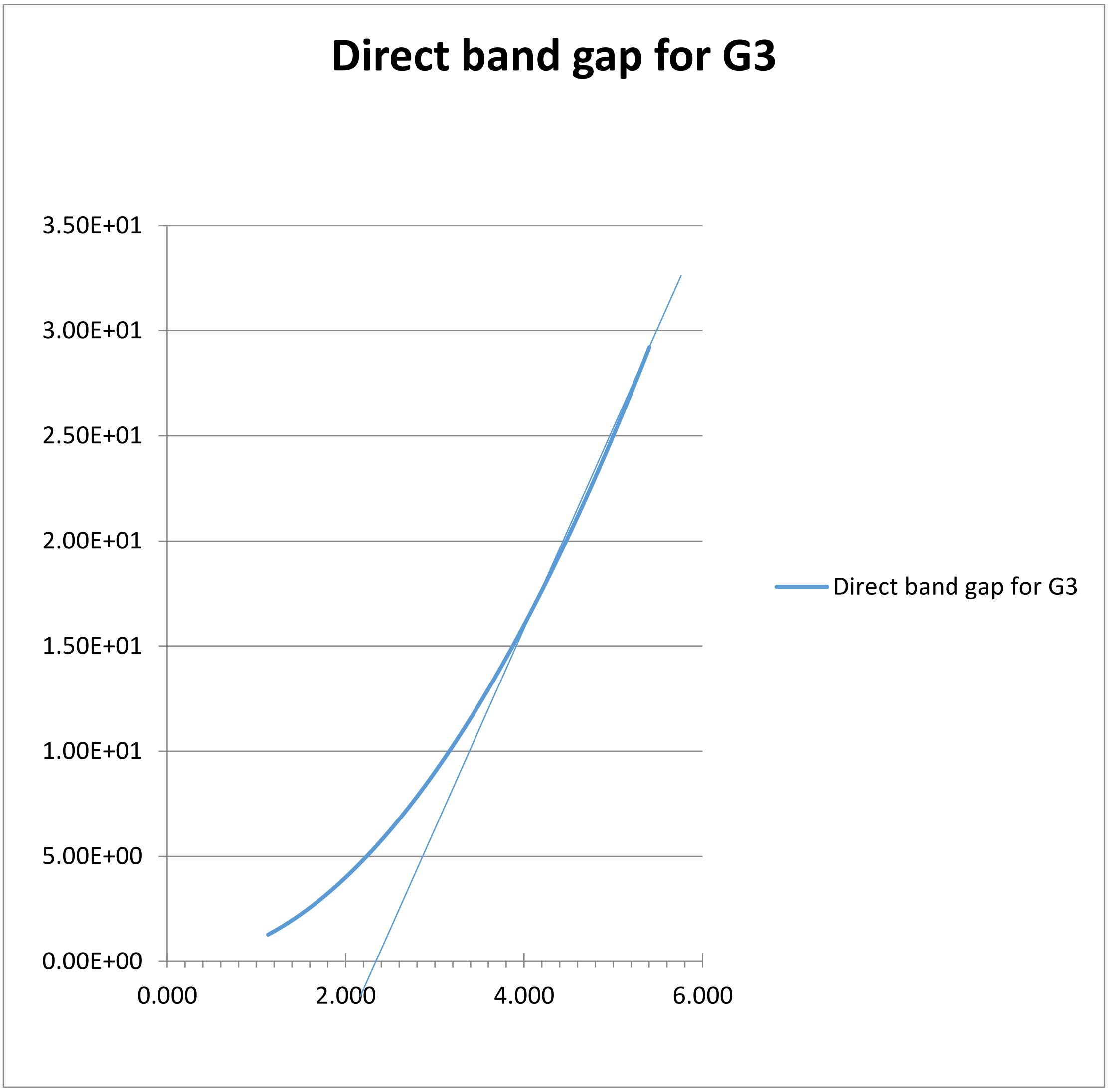

Figure 12. bandgap for sample G3 (7 layers)

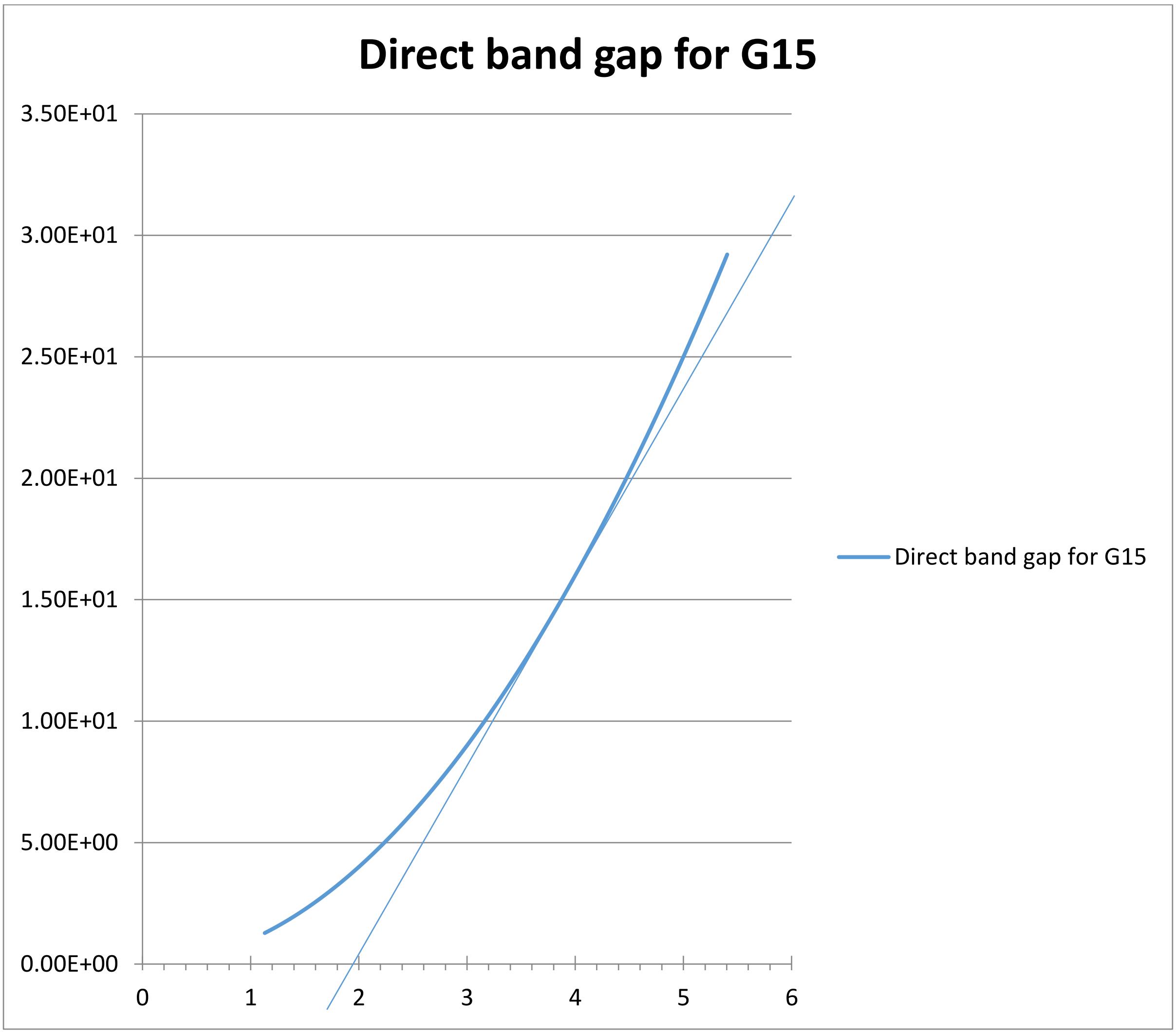

Figure 13. bandgap for sample G15 (9 layers)

The direct optical band gap for the samples was estimated to be G15 = 2.0eV, G3 = 2.4eV, G12 = 2.1eV, G13 = 2.3eV, and G7 = 2.2eV by extrapolating the linear portion of the curve $(h\nu)^2$ against $(h\nu)$ which is in line with results reported by Amaechi and Ogbonda (2022).

Conclusion

The advent of Graphene has significantly influenced the scientific realm and is poised to emerge as a ubiquitous material within the electronics community shortly. To achieve effective production, it is important to create graphene sheets on a large scale while considering their band gap, conductivity, resistivity, and transmittance properties. There has been a notable increase in the availability and utilisation of ecologically sustainable reductants in the process of synthesising graphene materials. The fabrication of thin films of graphene oxide has been effectively achieved by the utilisation of the modified Hummer process. Moreover, the samples must undergo an annealing process to transform into reduced graphene oxide. The effective preparation of the films was demonstrated through the synthesis, profilometry, and optical characterisation.

Recommendation

Graphene should be used as a substitute for Platinum as graphene can be easily produced which could be used as a counter electrode due to their high level of lucidity and excellent electrocatalytic activity.

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