Synthesis of Graphene through Electrochemical Exfoliation of Sri Lankan Graphite

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Abstract

Graphene, a remarkable two-dimensional carbon allotrope characterized by a hexagonally arranged carbon lattice, has garnered significant attention due to its extraordinary properties and diverse range of applications. For the synthesis of graphene, multiple methods are available. In this research, we opted for the electrochemical exfoliation method due to its simplicity, scalability, and environmentally friendly attributes. This methodology follows a top-down paradigm, whereby graphene is derived from graphite. The experimental configuration involved the construction of an electrolytic cell, employing carbon electrodes fabricated from compacted graphite powder, with a 0.1M Na_2SO_4 solution serving as the electrolyte. By systematically varying the voltage, current, and spatial separation between the anode and cathode, five experimental trials were conducted. Subsequently, the electrolyte underwent filtration, and the resultant residue underwent a drying process. Morphological observation of the synthesized graphene samples was facilitated using scanning electron microscopy (SEM). Furthermore, the confirmation of graphene sample purity was achieved through energy dispersive x-ray spectroscopy (EDS). The x-ray diffraction (XRD) analysis revealed a distinct diffraction peak at $2\theta=26.4°$, corresponding to the (002) plane. Additionally, the absorption peak of graphene was identified at 230 nm. Our findings strongly suggest that electrochemical exfoliation represents a promising avenue for the synthesis of graphene utilizing Sri Lankan graphite. However, further investigations are imperative to refine and optimise this method for the large-scale production of graphene.

Keywords: Graphite powder, Pressed graphite electrodes, SEM, Sodium sulphate, XRD

1 Introduction

Graphene is a two-dimensional carbon material with a hexagonal lattice structure [1], first isolated in 2004 by Andre Geim and Konstantin Novoselov, earning them the Nobel Prize in Physics in 2010. Various industries can benefit greatly from graphene's outstanding mix of thermal [2] electrical [3], optical [4], and mechanical [5], qualities. The unparalleled electrical conductivity and atomic-scale thinness of graphene in the domain of electronics hold the prospect of facilitating faster and more efficient device functionalities. Owing to its exceptional mechanical strength, graphene emerges as a compelling choice for advanced materials such as flexible

electronics and highly robust composites [6]. Moreover, improvements in energy storage [7], sensors, and even medical applications [8] are made possible by its excellent thermal conductivity and transparency. Graphene is an abundant and adaptable substance that has the potential to revolutionize a variety of global businesses and technological fields.

The synthesis of graphene involves two primary methods. The top-down method involves reducing bulk graphite through mechanical, chemical, or electrochemical means into thin graphene layers, and the bottom-up method, creates graphene atom by atom or molecule by molecule through chemical reactions [9]. The top-down approach produces graphene from graphite using processes including mechanical exfoliation [1] and chemical oxidation [10], which is simple but may compromise quality and scalability. The bottom-up approach, demonstrated by chemical vapor deposition [11] and epitaxial growth, assembles carbon atoms or molecules into exact graphene structures, assuring excellent quality and customized features, but often at higher costs. The preferred graphene characteristics, scalability, and planned applications will influence the technique selection.

Despite its enormous potential, difficulties with large-scale production and integration with current technology persist, necessitating more study and development. Nonetheless, graphene's properties make it a promising material with a wide array of applications in modern science and technology.

Electrochemical exfoliation is a promising technique for producing single or few-layer graphene from bulk graphite through an electrochemical process [12]. By immersing a graphite electrode in an electrolyte solution and applying an electrical potential, graphene layers exfoliate, resulting in graphene suspended in the solution [13]. Factors, like applied electrical potentials, currents, processing time, and electrolyte composition, influence the quality of the produced graphene [14].

The chosen approach of electrochemical exfoliation is particularly promising, offering numerous advantages, such as its simplicity, scalability, and ability to preserve the intrinsic properties of the starting graphite material.

The history and geology of graphite in Sri Lanka reveal its significance as a valuable mineral resource [15]. Graphite, known as "plumbago" and "black lead" in the past, has been used for centuries to make pencils. Sri Lanka has a long tradition of graphite mining, with trade dating back to the 16th century. Notably, graphite occurs in three main types: flake, vein, and amorphous, with vein graphite being the most economically significant [16]. The geology and structure of these deposits involve various rock types, and the graphite veins show different crystalline forms [17]. The country's graphite deposits are concentrated in the central belt, contributing significantly to the economy and global graphite trade [18].

This research primarily explores the potential for producing graphene by electrochemically exfoliating graphite sourced from Sri Lanka. The process involves utilizing pressed graphite electrodes made from graphite powder.

2 Methodology

To begin the process, high-quality (+99% carbon content) natural crystalline graphite powder was carefully selected as the raw material. These graphite samples were obtained from Bogala Graphite Lanka PLC.

The collected graphite samples underwent characterization to assess their physical and chemical properties. Scanning

Electron Microscopy (SEM) and Energy Dispersive X-ray Spectroscopy (EDS) were employed to examine the crystal structure, purity, and morphology of the graphite powder.

The electrode preparation followed, involving the custom-made fabrication of both the anode and cathode electrodes from graphite powder without using any binder material. The graphite powder was sieved to achieve a uniform particle size distribution, and particles passing through a 75-micron sieve were selected for electrode production. A mold was then utilized to shape the graphite powder into the desired electrode form. The electrodes, as depicted in Fig. 1, were produced in a rectangular shape with dimensions of 95mm×45mm×3mm (length × width × thickness). This was achieved by applying a pressure of 6500 psi (44.8MPa) using a hydraulic press.

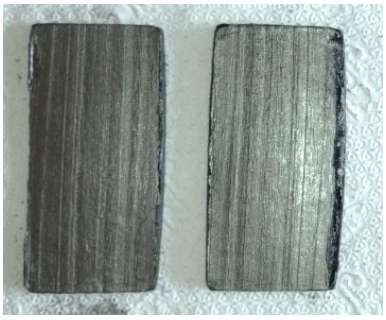

Figure 1: Pressed graphite electrodes

Following the electrode preparation, the selection and preparation of the electrolyte solution were carefully carried out. A 0.1 M sodium sulfate (Na_2SO_4) solution was chosen as the electrolyte due to its conductivity for the exfoliation process and compatibility with the graphite electrodes [14]. To prepare the solution, 14.204 grams of sodium sulfate powder were accurately weighed using an analytical balance and then dissolved in a 1000ml volumetric flask containing deionized water.

Next, the electrolytic cell was arranged as indicated in Fig. 2, serving as the platform for the electrochemical exfoliation process. The rectangular glass container, chemically inert and with dimensions of 150 mm × 100 mm × 100 mm (length × width × height), was fabricated for this purpose. Two stands made of Styrofoam were used to position the electrodes inside the electrolytic cell, allowing adjustment of the distance between the two electrodes. The anode and cathode electrodes, fabricated using pressed graphite powder, were carefully placed inside the cell, and crocodile clips were attached to the electrodes using copper plates to ensure better electrical contact.

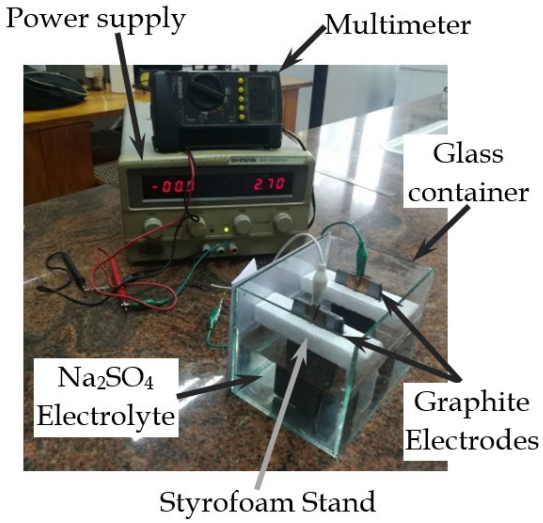

Figure 2: Electrolytic cell used for the test

The electrochemical exfoliation process was initiated by applying a controlled voltage between the two graphite electrodes immersed in the electrolyte solution. The process was carried out by maintaining a specific voltage value for a test and a defined exfoliation time, to achieve desired results. The electrical potential was turned off once the predetermined amount of time had passed, and the electrodes were then washed and taken off. The electrolyte was filtered, and the residue was collected and dried.

By changing the parameters, five tests were carried out as in Table 1.

Table 1: Parameters used in the tests

No	Voltage [V]	Current density [mA/mm²]	Electrode distance [cm]	Time [min]
T1	8.2	0.13	7.5	10
T2	7.1	0.14	5.0	10
T3	5.1	0.04	5.0	10
T4	5.0	0.09	7.5	10
T5	5.0	0.13	3.0	10

Finally, the filtered and dried powder underwent a characterization step to assess its structural and morphological properties. SEM and EDS analysis were done utilizing ZEISS EVO 18 Research. XRD analysis was carried out using Bruker D8 Advance ECO. The UV-Visible spectra of graphene sample were tested using Thermo Scientific GENESYS 10S UV Vis Spectrophotometer.

3 Results

3.1 SEM and EDS analysis

Scanning Electron Microscopy (SEM) was employed for high-resolution examination of the surface morphology [19]. Energy dispersive x-ray analysis (EDS) was performed to confirm the purity of the sample and identify any potential impurities or contaminants present [20].

The SEM analysis was conducted on powdered natural crystalline graphite samples, and the corresponding image is presented in Fig. 3(a). Additionally, Table 2 provides the findings from the EDS analysis of the graphite sample.

Table 2: The weight and atomic percentage of graphite powder

Element	Weight %	Atomic %
C	99.64	99.84
O	0.01	0.01
Si	0.35	0.15
Total	100.00	100.00

Fig. 3(b) illustrates the SEM image of a graphene sample. The purity of the exfoliated graphene sample was observed through EDS spectrum in Fig. 4, indicating carbon as the predominant element, constituting 99.64% by weight in the sample. The atomic and weight percentages of the elements in the exfoliated graphene are detailed in Table 3.

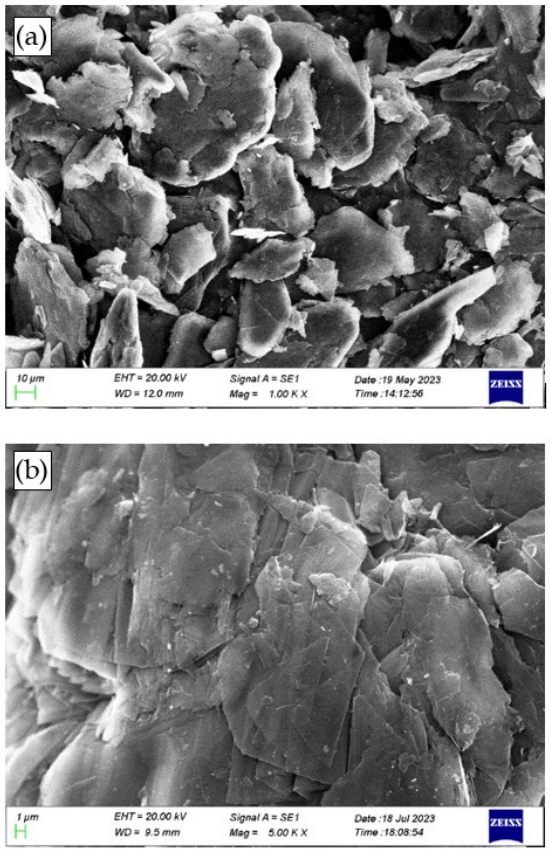

Figure 3: SEM images of (a): the graphite powder (b): graphene sample from T2

Table 3: The weight and atomic percentage of exfoliated graphene sample T2

Element	Weight %	Atomic %
C	98.58	98.99
O	1.22	0.92
Si	0.19	0.08
Total	99.99	99.99

3.2 XRD

The X-ray diffraction (XRD) analysis presents the distinct diffraction patterns corresponding to the crystallographic structure [20]. As in Fig. 5, the XRD

spectrum displayed a prominent reflection peak at 2θ=26.4°.

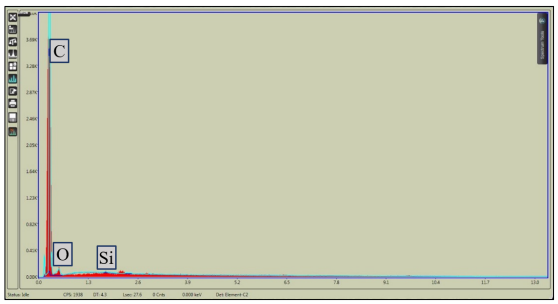

Figure 4: EDS spectrum of graphene sample T2

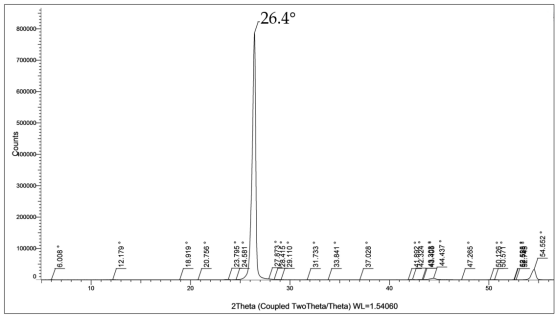

Figure 5: XRD test result of graphene sample T_3

3.2 UV vis spectroscopy

The UV-Visible (UV-Vis) spectrum analysis of the graphene sample exhibited a notable absorption peak at around 230 nm (Fig. 6).

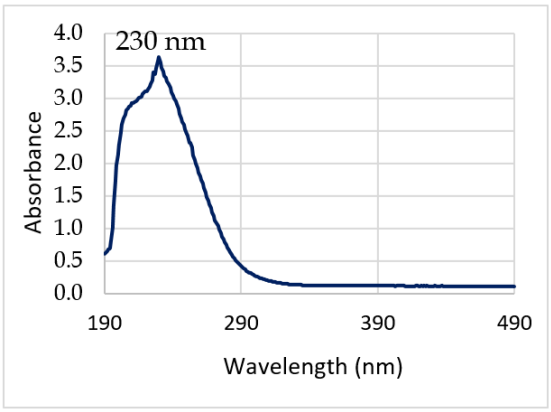

Figure 6: Absorption spectrum of graphene sample T4

4 Discussion

The electrochemical method to make graphene from graphite is cost-effective, scalable, and versatile. It's a more economical choice compared to expensive methods like chemical vapour deposition (CVD) [11], making it feasible for large-scale graphene production. Operating at room temperature reduces energy use, adding to its cost-effectiveness. The simple setup makes it easily scalable for industry use [21], potentially bringing graphene to various sectors.

In the study, SEM analysis was conducted to understand the microstructure and particle characteristics of graphite powder and exfoliated graphene sample.

The image of the graphite powder revealed a flake-like structure with multiple layers stacked together, typical of graphite. The graphite sample's elemental content was assessed through EDS analysis.

The SEM image of the synthesized graphene sample confirms the formation of graphene layers [22]. However, the graphene sheets are not perfectly flat but exhibit crumpled surfaces and out-of-plane deformation, resembling a crinkly/wrinkly paper-like appearance [23]. These surface roughness features arise due to the deformation occurring during the exfoliation and restacking processes [13]. Nonetheless, these characteristics are typical of graphene and validate its formation. The image of the graphene powder illustrates random sizes and shapes, indicating a rippled and uneven structure [24], which is a consequence of the stability of graphene during the electrochemical exfoliation process [25].

The EDS analysis of the graphene sample was performed to determine its elemental composition. The EDS spectrum revealed prominent peaks corresponding to carbon (C), oxygen (O) and silicon (Si) elements. The strong carbon peak is indicative of the high carbon content in the sample. The absence of peaks corresponding to other elements further supports the less impurity content of the sample [26]. However,

additional tests are necessary to validate these results due to the inherent limitations of EDS.

The XRD analysis was performed to study the crystal structure of the exfoliated graphene sample. The XRD spectrum showed a reflection peak at 2θ=26.4°, corresponding to the (002) plane, indicating its crimped structure and increased interlayer spacing [13]. This confirmed the formation of graphene through the process [27]. The XRD analysis verified the production of graphene and provided structural insights.

From the UV-Visible spectrum, it was observed that the absorbance is at its maximum at 230 nm, which corresponds to the p - p transition of C - C bonds [28].

5 Conclusions

The study focused on synthesizing graphene through electrochemical exfoliation of Sri Lankan graphite powder. An electrolytic cell was designed to facilitate the exfoliation process using pressed graphite plates as electrodes and a 0.1M Na_2SO_4 solution as the electrolyte. Five tests were conducted with varying parameters.

The results from SEM and EDS analysis confirmed the exfoliation of graphite into graphene layers. The SEM images displayed crumpled and uneven surfaces, characteristic of graphene, validating its formation. EDS analysis provides an indication of the purity of the synthesized graphene sample, revealing a dominant carbon content.

XRD analysis provided structural insights, showing a reflection peak at 2θ=26.4° corresponding to the (002) plane of graphene. This peak broadening indicated the crimped structure and increased interlayer spacing, supporting the formation of graphene. Additionally, UV-Visible spectroscopy demonstrated that the synthesized graphene exhibited absorbance at 230 nm, corresponding to the p-p transition of C-C bonds [28].

Through this study, the electrochemical exfoliation method proved to be a favourable route for synthesizing graphene from Sri Lankan graphite. Further research is required to enhance this method for large-scale production of graphene, opening new possibilities for its applications in various fields.

Acknowledgements

The authors would like to express gratitude to Bogala Graphite Lanka PLC for supplying of graphite samples required for the study. Also, authors would like to express their thank to the Head and staff of the Materials Science and Engineering Department at the University of Moratuwa for their invaluable support in conducting essential tests. Additionally, thanks to all individuals who offered their assistance and inspiration throughout this research endeavor.

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