UDC 621.315.592:546.26:66.081.63

--------MATERIALS SCIENCE

-0

The object of this study is a graphene-like material synthesized from coconut shell biomass via a solvothermal process using ethylene glycol (98%) as the dispersing medium. Were examined are exfoliated carbon nanosheets intended for use as electrode materials in supercapacitors. The research addresses the problem of improving biomass-derived carbon materials' structural quality and electrochemical performance for energy storage. The solvothermal process was applied at varying ethylene glycol concentrations (1, 3, and 5 mg/ml), and the synthesized samples were compared with an untreated control. The results show that the sample treated with 3 mg/ml (SL-2) exhibited the most favorable characteristics, including reduced interlayer spacing (0.39 nm), formation of thin nanosheets, and decreased oxygen-containing functional groups, as evidenced by TEM, FTIR, and EDS analyses. These structural improvements are attributed to the combined effects of thermal energy and solvent-assisted exfoliation, which facilitated partial deoxygenation and reordering of carbon layers. BET analysis revealed a high specific surface area of 872.886 m²/g, contributing to enhanced ion accessibility. Electrochemical measurements demonstrated a specific capacitance of 31.50 F/g for SL-2, significantly higher than the untreated sample (6.32 F/g), along with lower internal resistance (1.87 Ω) and prolonged charge-discharge time (39.90 s), indicating improved ion transport and conductivity. These results highlight the potential of this sustainable and tunable method for producing cost-effective, eco-friendly supercapacitor electrodes

Keywords: graphene-like material, coconut shell biomass, solvothermal treatment, ethylene glycol, electrochemical performance, supercapacitor

-0 -

DOI: 10.15587/1729-4061.2025.330856 **IDENTIFYING THE INFLUENCE OF** SOLVOTHERMAL PROCESSING **ON THE STRUCTURAL AND ELECTROCHEMICAL PROPERTIES OF GRAPHENE-LIKE MATERIALS FOR SUPERCAPACITOR APPLICATIONS** Wahyu Widanarto Corresponding author Doctor of Engineering Sciences* E-mail: wahyu.widanarto@unsoed.ac.id Dedi Setiawan Bachelor of Engineering Science* Mukhtar Effendi Doctor of Engineering Sciences* Wahyu Tri Cahyanto

Retno Supriyanti

PhD*

Doctor of Engineering Sciences**

Muhammad Syaiful Aliim

Master of Engineering Sciences**

Dina Rahmawati

Master of Engineering Sciences Department of Industrial Engineering **Telkom University**

Dl. Panjaitan str., 128, Purwokerto, Jawa Tengah, Indonesia, 53147

Candra Kurniawan

Doctor of Engineering Sciences Research Center for Advanced Materials Badan Riset dan Inovasi Nasional Puspiptek Office Area, 224, South Tangerang, Indonesia, 15314

*Department of Physics

Universitas Jenderal Soedirman

DR. Soeparno str., 61, Karangwangkal, Purwokerto Utara, Banyumas, Jawa Tengah, Indonesia, 53123

Kurniawan, C. (2025). Identifying the influence of solvothermal processing on the structural and electrochemical prop-

erties of graphene-like materials for supercapacitor applications. Eastern-European Journal of Enterprise Technologies,

**Department of Electrical Engineering Universitas Jenderal Soedirman

3 (12 (135)), 6-14. https://doi.org/10.15587/1729-4061.2025.330856

Mayjend Sungkono str., 5, Purbalingga, Indonesia, 53371

How to Cite: Widanarto, W., Setiawan, D., Effendi, M., Cahyanto, W. T., Supriyanti, R., Aliim, M. S., Rahmawati, D.,

Received 01.04.2025 Received in revised form 20.05.2025 Accepted 09.06.2025 Published 25,06,2025

1. Introduction

The global demand for fast, efficient, and sustainable energy storage technologies is rapidly increasing, driven by the proliferation of portable electronics, electric vehicles, and integrating renewable energy into modern power grids. Supercapacitors have emerged as one of the most promising solutions due to their high power density, rapid charge-discharge capabilities, long cycle life, and relatively low production costs. These features make them well-suited for bridging the gap between conventional capacitors and batteries, particularly in applications requiring high power and reasonable energy density [1].

To achieve optimal performance, supercapacitor electrodes must exhibit a large specific surface area, appropriate pore distribution to enable electrolyte ion diffusion, and excellent electrical conductivity to facilitate rapid charge transport [2]. Among various materials studied for this purpose, carbon-based materials – especially graphene – have gained significant attention due to their outstanding electrical, mechanical, and chemical properties. Graphene is a two-dimensional (2D) material composed of sp²-hybridized carbon atoms in a hexagonal lattice, which gives rise to its high electrical conductivity, large surface area, and mechanical strength [3]. These characteristics make graphene a leading candidate for next-generation electrochemical energy storage systems.

However, conventional graphene synthesis techniques – such as chemical vapor deposition (CVD), graphite exfoliation, and chemical oxidation – suffer from limitations, including high cost, complex processing, and environmental concerns related to hazardous chemical use [4]. As a result, there is a growing shift towards green and cost-effective synthesis methods using biomass-derived carbon precursors, such as rice husks, peanut shells, wheat straw, sawdust, palm oil waste, and coconut shells [5]. Coconut shells, particularly, are attractive due to their high carbon content, widespread availability, and potential for conversion into porous carbon materials through carbonization and chemical activation using KOH [5].

Despite the effectiveness of these methods in producing activated carbon, the resulting materials often lack the desired structural features of graphene. The solvothermal process has recently gained traction as a post-treatment strategy to improve graphitization and induce graphene-like properties. Organic solvents such as ethylene glycol function as reducing agents and dispersion media, promoting exfoliation and rearrangement of carbon sheets into thin-layered, high surface area structures with dominant sp² bonding characteristics [6]. These graphene-like materials share functional similarities with pristine graphene, although they may differ in structural perfection, defect density, and surface chemistry.

Nevertheless, one underexplored aspect in this field is how variations in solvent concentration during solvethermal treatment influence the transformation of biomass-derived carbon into graphene-like materials. Addressing this knowledge gap is crucial for optimizing the synthesis parameters and tailoring material performance for supercapacitor applications.

Therefore, research on developing graphene-like materials from coconut shell biomass via solvothermal processing is highly relevant. It responds to the growing scientific and technological demand for environmentally sustainable, high-performance carbon materials in electrochemical energy storage systems.

2. Literature review and problem statement

Recent advancements in electrochemical energy storage have underscored the importance of developing efficient, cost-effective, and sustainable electrode materials. Carbon-based materials, particularly graphene, and its derivatives, have been extensively investigated due to their high

surface area, excellent electrical conductivity, and chemical stability [3]. Two-dimensional materials synthesized through layer-by-layer approaches have shown significant potential for energy applications owing to their tunable morphology and functional properties [7].

Despite these advantages, conventional graphene synthesis methods such as chemical vapor deposition, mechanical exfoliation, and epitaxial growth face scalability limitations, high energy requirements, and dependence on expensive equipment [4, 8]. While practical, chemical approaches like the Hummers method involve toxic oxidizing agents and generate hazardous waste, raising environmental concerns for large-scale production [9]. These challenges are compounded by the inherent complexity of biomass composition, low graphitization degrees, and difficulties in precisely controlling porosity and microstructure at the nanoscale when using biomass-derived precursors.

To address these issues, biomass-derived carbon sources such as coconut shells, rice husks, and banana peels have been explored as sustainable alternatives [5, 10, 11]. Among these, coconut shells are particularly promising due to their high fixed carbon content and low ash levels. However, transforming these precursors into graphene-like materials with optimal electrochemical properties remains a significant challenge. Solvothermal processing has emerged as a potential solution, enabling improved exfoliation and structural modification. For example, nitrogen-doped reduced graphene oxide has been synthesized via solvothermal methods, though the influence of solvent concentration was not examined [6]. Similarly, graphene and reduced graphene oxides have been produced from coconut shell biomass without comprehensive electrochemical analysis or optimization of process parameters such as solvent concentration [12, 13].

These limitations highlight unresolved questions regarding the role of solvent conditions in determining the morphology, surface area, and electrochemical performance of biomass-derived graphene-like materials. Specifically, the impact of ethylene glycol concentration during solvothermal processing has not been systematically investigated, leaving a critical gap in the development of scalable and environmentally friendly synthesis routes. All this allows to assert that it is expedient to conduct a study on the influence of ethylene glycol concentration during solvothermal treatment on the structural and electrochemical properties of graphene-like materials derived from coconut shell biomass, particularly in the context of their application as supercapacitor electrodes.

3. The aim and objectives of the study

The aim of the study is to identifying the influence of solvothermal processing using ethylene glycol (98%) as the dispersing medium on the structural and electrochemical properties of graphene-like materials derived from coconut shell biomass, particularly in the context of their application as supercapacitor electrodes. This will allow the identification of optimal processing conditions for producing high-performance, sustainable electrode materials, which can be practically applied in the development of cost-effective and environmentally friendly energy storage devices.

To achieve this aim, the following objectives are accomplished:

- to identify the structural, morphological, and chemical properties of the graphene-like materials using X-ray diffrac-

tion (XRD), transmission electron microscopy (TEM HRTEM), Fourier-transform infrared spectroscopy (FTIR), and Brunauer-Emmett-Teller (BET) surface area analysis;

– to evaluate the electrochemical performance of the materials using cyclic voltammetry (CV), galvanostatic charge–discharge (GCD), and electrochemical impedance spectroscopy (EIS), in order to determine the optimal dispersion condition that enhances specific capacitance, reduces internal resistance, and improves charge–discharge behavior.

4. Materials and methods

The object of this study is a graphene-like material synthesized from coconut shell biomass via a solvothermal process using ethylene glycol (98%) as the dispersing medium. The study hypothesizes that varying the concentration of ethylene glycol significantly affects the crystalline structure, surface morphology, chemical composition, and electrochemical performance of the resulting material. It is assumed that ethylene glycol acts not only as a dispersion medium but also as a structure-directing agent under solvothermal conditions. For simplification, the influence of other synthesis parameters such as reaction time, temperature, and biomass pretreatment was held constant to isolate the effect of ethylene glycol concentration.

This study was carried out experimentally using a biomass-based material synthesis approach. Coconut shells were cut into pieces measuring 3-6 cm, cleaned from fibrous husk, washed with 98% distilled water, and dried at 105°C for 2 hours using a laboratory oven (Mummert Oven UN 53 L). The dried shells were then carbonized and chemically activated using a 90% KOH solution with a mass ratio of 1:4 (carbon: KOH) to enhance surface area and develop porous structures. The resulting activated carbon was rinsed with distilled water until neutral pH and subsequently dried at 100°C for 24 hours. The synthesis of activated carbon was carried out following a modified procedure previously developed by our research group [5]. The dry carbon powder was dispersed in 98% ethylene glycol at concentrations of 0, 1, 3, and 5 mg/ml and stirred using a magnetic hotplate stirrer (Thermo Scientific SP88857105) at 500 rpm for 4 hours to ensure homogeneous mixing. The dispersed samples were labelled SL-0 (no ethylene glycol), SL-1 (1 mg/ml), SL-2 (3 mg/ml), and SL-3 (5 mg/ml), respectively. Each sample was transferred into a 100 ml Teflon-lined stainless-steel autoclave and subjected to solvothermal treatment at 180°C for 16 hours using a muffle furnace (PPF-1300, LIPI). This process aimed to evaluate the effect of ethylene glycol concentration on the reduction of oxygen-containing functional groups and the formation of graphene-like layered structures.

Characterization of the synthesized materials was conducted using various analytical instruments. The crystalline structure was identified by X-ray Diffraction (XRD, SmartLab 3 kW, Rigaku), while functional groups were determined using Fourier Transform Infrared Spectroscopy (FTIR, ThermoScientific Nicolet iS-10). Surface morphology and elemental composition were analyzed using Field Emission Scanning Electron Microscopy with Energy Dispersive Spectroscopy (FESEM-EDS, JIB-4610F), and the internal layered structure was observed through Transmission Electron Microscopy and High-Resolution TEM (TEM/HRTEM, FEI Tecnai G2 Supertwin). Specific surface area and pore size distribution were measured using the Brunauer-Emmett-Teller (BET) method with a Quan-

tachrome 11.03 surface area analyzer. The electrochemical performance was evaluated using a Corrtest electrochemical workstation by means of cyclic voltammetry (CV), galvanostatic charge–discharge (GCD), and electrochemical impedance spectroscopy (EIS) in a three-electrode system comprising carbon paste as the working electrode, Ag/AgCl as the reference electrode, and platinum wire as the counter electrode in 1 M $\rm Na_2SO_4$ electrolyte solution. All experimental procedures were conducted under consistent and controlled conditions to ensure the reproducibility and reliability of the results.

5. The results of studying the properties of graphenelike materials synthesized from coconut shell biomass via solvothermal treatment with varying ethylene glycol concentrations

5. 1. The study results on determining the structural, chemical, and morphological characteristics of graphene-like materials

The structural characteristics of the synthesized graphene-like materials in this study were examined using X-ray diffraction (XRD). The resulting diffraction patterns are presented in Fig. 1. The XRD profiles of samples prepared with 0, 1, 3, and 5 mg/l dispersion concentrations reveal their crystalline features and phase composition.

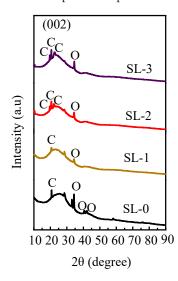


Fig. 1. Crystallinity and phase composition of graphene-like materials at various dispersion concentrations (0, 1, 3, and 5 mg/ml)

The chemical characteristics of the graphene-like materials were analyzed using Fourier-transform infrared spectroscopy (FTIR). The FTIR spectra obtained for samples prepared with dispersion concentrations of 0, 1, 3, and 5 mg/ml are presented in Fig. 2. The figure shows the absorption bands corresponding to various functional groups present in the materials.

The elemental composition of the graphene-like materials was examined using energy-dispersive X-ray spectroscopy (EDX). Fig. 3 presents the EDS spectra of samples synthesized at different ethylene glycol dispersion concentrations, showing the presence and relative abundance of constituent elements in the materials.

The surface morphology of the synthesized graphene-like materials was analyzed using scanning electron microsco-

py (SEM). Fig. 4 shows SEM micrographs of the samples prepared with varying dispersion concentrations. The images display each sample's surface texture and morphological features.

Transmission electron microscopy (TEM) and high-resolution TEM (HRTEM) analyses were performed to observe the internal structure and layer arrangement of the selected graphene-like samples. Fig. 5, a presents representative TEM images of SL-0 and SL-2 samples, Fig. 5, b presents representative HRTEM images of SL-0 and SL-2 samples highlighting differences in layer orientation and nanosheet formation.

Fig. 6 presents the nitrogen adsorption-desorption isotherms of the graphene-like materials synthesized at different dispersion concentrations, illustrating each sample's surface area behavior. Table 1 provides the specific surface area values derived from the isotherms using the BET method.

Fig. 7 presents the pore size distribution of graphene-like materials as determined by the BJH method based on nitrogen desorption data.

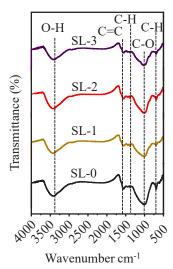


Fig. 2. Identification of functional groups in graphene-like materials at various dispersion concentrations

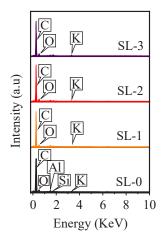


Fig. 3. Elemental composition of graphene-like materials synthesized at different dispersion concentrations

Thus, the obtained data (Fig. 1–7 and Table 1) reflect the graphene-like materials synthesized at various dispersion concentrations' structural, chemical, and morphological characteristics. The differences in crystallinity, surface morphology, porosity, and functional group composition across the samples were experimentally established.

Table 1
The specific surface area of the studied samples

Sample code	Slope (s)	Intercept (i)	The volume of nitrogen gas (V_m)	Specific surface area $S_{\rm BET}$ (m ² /g)
SL-0	2.909	-0.01053	0.345	1201.710
SL-1	5.281	0.01466	0.188	657.570
SL-2	3.694	0.02570	0.268	872.886
SL-3	5.721	0.06569	0.173	601.815

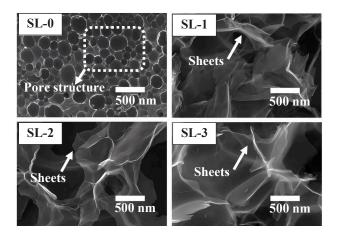


Fig. 4. Surface morphology of graphene-like materials synthesized at various dispersion concentrations

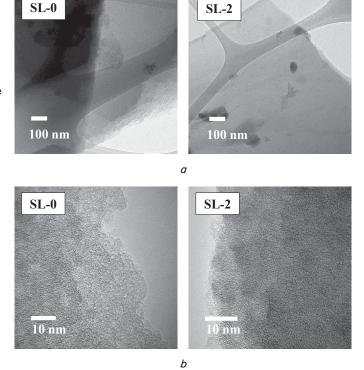


Fig. 5. Internal structure and layer arrangement of selected graphene-like samples (SL-0 and SL-2):

a —Transmission Electron Microscopy (TEM);

b—High-Resolution Transmission Electron Microscopy (HRTEM)

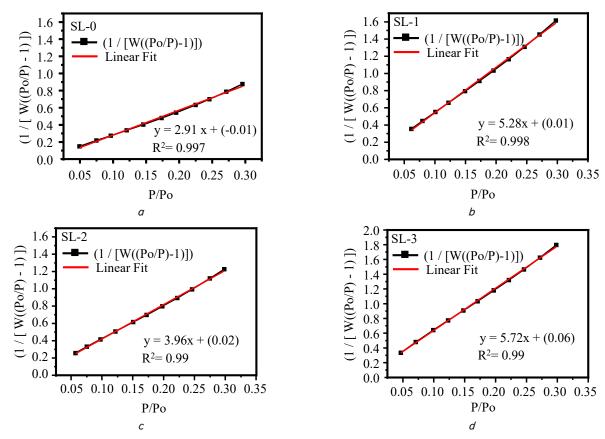


Fig. 6. Nitrogen adsorption-desorption isotherms of graphene-like materials prepared with different dispersion concentrations: α – SL-0; b – SL-1; c – SL-2; d – SL-3

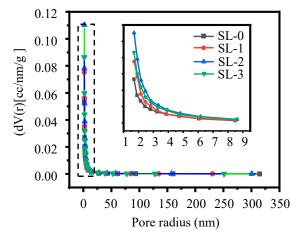


Fig. 7. Pore size distribution of graphene-like materials

5. 2. The study results on determining the electrochemical properties of selected graphene-like materials

The charge storage behavior of the selected graphene-like materials (SL-0 and SL-2) was evaluated through cyclic voltammetry analysis. Fig. 8 depicts the voltammograms recorded at a scan rate of 10~mV/s.

Using a galvanostatic technique, the charge-discharge characteristics of SL-0 and SL-2 were examined under different current densities. Fig. 9 displays the corresponding charge-discharge curves obtained from this measurement.

Fig. 10 shows Nyquist plots of SL-0 and SL-2 samples obtained from electrochemical impedance spectroscopy (EIS) measurements. The curves indicate internal resistance and ion diffusion behavior.

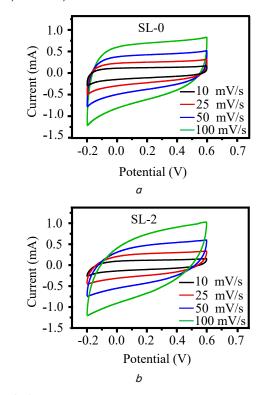


Fig. 8. Charge storage behavior of graphene-like materials analyzed via cyclic voltammetry under various scan rates: a-SL-0; b-SL-2

The experimental data shown in Fig. 8–10 provide information on the electrochemical responses of SL-0 and SL-2 samples.

These include their capacitance behavior, charge-discharge performance, and internal resistance characteristics as determined from cyclic voltammetry, galvanostatic cycling, and EIS tests.

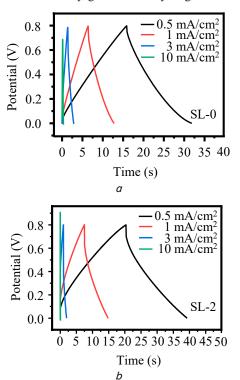


Fig. 9. Charge-discharge characteristics of graphene-like materials under various current densities: a - SL-0; b - SL-2

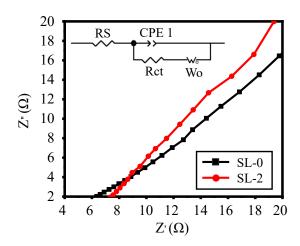


Fig. 10. Electrochemical impedance behavior of SL-0 and SL-2 samples indicating internal resistance and ion diffusion

6. Discussion of the study results on determining the structural, chemical, morphological, and electrochemical characteristics of graphene-like materials

Fig. 1 presents the XRD patterns of graphene-like materials synthesized through solvothermal treatment with varying concentrations of ethylene glycol dispersion. The broad hump around 25–26° is characteristic of the (002) peak of graphene-like materials, indicating the presence of thin carbon layers. In the sample without solvothermal treatment (SL-0), additional peaks were observed between 30° and 45°, associated with re-

sidual functional groups such as carboxylate, as confirmed by FTIR results. After the solvothermal treatment, the intensity of peaks associated with oxygen content between 30° and 45° decreased, indicating the reduction of residual functional groups such as carboxylate due to the solvothermal reduction process. It is supported by the FTIR characterization results, where solvothermal-treated samples (SL-1, SL-2, and SL-3) exhibit a reduction in absorption intensity for functional groups such as C-H stretching (alkane), C-O stretching, and C-H stretching (alkene). Additionally, the SL-0 sample exhibited an average interlayer spacing (d-spacing) of 0.57 nm.

In contrast, the SL-2 sample, which underwent solvothermal treatment with a 3 mg/ml dispersion concentration, showed a reduced interlayer spacing of 0.39 nm, consistent with the values obtained from HRTEM analysis. The change in interlayer spacing after the solvothermal process suggests that thermal reduction leads to layer separation and the elimination of oxygen functional groups, such as hydroxyl, carboxylate, and epoxy, which were previously intercalated between the crystal layers [14, 15]. FTIR results further support it, which show a decrease in the absorption intensity of functional groups such as O-H, C-O, and C-H stretching in the solvothermal-treated samples (SL-1, SL-2, and SL-3). These findings are consistent with previous studies [16]. Among all variations, the SL-2 sample with 3 mg/ml ethylene glycol dispersion exhibited the most significant structural changes, characterized by forming thin carbon layers and a substantial reduction in oxygen content. These findings will be elaborated in the following sections.

Fig. 2 presents the FTIR spectroscopy results of the graphene-like materials, highlighting several key functional groups. The absorption band observed at 3417 cm⁻¹ corresponds to the O-H stretching functional group, while the peak at 1556 cm⁻¹ is associated with the C=C functional groups, characteristic of aromatic bonds. Additionally, wavenumbers at 1383, 1028, and 695.44 cm⁻¹ reflect the C-H stretching of alkanes, C-O stretching, and C-H stretching of alkenes, respectively. The FTIR spectrum effectively identifies the functional groups in each sample, from SL-0 to SL-3, demonstrating that aromatic C=C functional groups dominate the structure of the graphene material, with electrons in the π bonds generally delocalized across the aromatic ring [17]. After solvothermal reduction, namely samples SL-1, SL-2, and SL-3, show a decrease in absorption intensity in the O-H, C-H (alkane), C-O, and C-H (alkene) groups, indicating the reduction of hydroxyl and epoxy groups. This decrease confirms that the solvothermal process effectively reduced most oxygen functional groups [18].

The EDS spectrum (Fig. 3) supports the FTIR results (Fig. 2), confirming carbon dominance and oxygen reduction after solvothermal treatment. The composition in the SL-0 sample, before solvothermal treatment, includes 93.7 at.% carbon and 4.7 at.% oxygen, consistent with O-H and C-O groups observed in the FTIR spectrum. After solvothermal treatment (SL-1, SL-2, SL-3), the carbon content significantly increased to 98.3–98.6 at.%, with a reduction in oxygen and other impurities. The presence of elements such as K, Si, and Al in the samples is likely derived from the mineral residues naturally present in coconut shells used as raw material. Coconut shells naturally contain mineral ash composed of various elements, including potassium (K), silicon (Si), and aluminum (Al). These elements can persist during the pyrolysis or solvothermal, especially if not completely removed during the washing or initial purification stages. The presence of K, Si, and Al can influence the properties of the resulting material. For instance, K can act as a catalyst during the carbonization process, affecting the crystal structure and

morphology of the material. Meanwhile, Si and Al, which are typically bound in the form of silicate or aluminosilicate compounds, can influence the thermal stability and adsorption capacity of the material due to the potential formation of additional pores. This reduction aligns with the decreased absorption intensity of oxygen-containing groups in the FTIR spectrum, indicating that the solvothermal process effectively reduces oxygen groups and increases carbon content, particularly in SL-2, which has the highest carbon content [19].

The surface morphology of graphene-like materials before and after solvothermal treatment at various dispersion concentrations was analyzed using FESEM. As shown in Fig. 4, the surface morphology of samples (SL-1, SL-2, and SL-3) after solvothermal treatment showed thin, crumpled sheet structures characteristic of graphene-like materials. These results are consistent with those reported by other researchers [20]. In contrast, samples that did not undergo solvothermal treatment displayed the formation of unevenly distributed porous structures. These findings confirm that solvothermal treatment significantly influences the morphology of the samples.

Fig. 5 illustrates the atomic structure images from TEM and HRTEM of samples prepared with and without the solvothermal process at a specific concentration. Based on the TEM images, the SL-0 sample exhibits a darker contrast, indicating a graphene-like structure with stacked nanosheets and thick graphene sheets of multiple layers. In contrast, the SL-2 sample shows a lighter contrast, suggesting a thinner nanosheet structure typical of graphene-like materials. The more transparent regions prove that few-layer graphene nanosheets were successfully exfoliated due to the solvothermal treatment. The HRTEM results showed that the SL-0 sample had an average interlayer spacing of 0.57 nm. After the solvothermal treatment at a concentration of 3 mg/ml, the SL-2 sample exhibited a reduced interlayer spacing of 0.39 nm. This reduction may indicate decreasing interlayer oxygen content and enhanced van der Waals interactions between the graphene sheets. Although transparency and interlayer spacing changes may indicate differences in layer number, a more quantitative analysis would require additional techniques such as Raman spectroscopy or AFM. Therefore, this interpretation is primarily based on morphological indications from TEM rather than an exact estimation of the layer number. These findings are consistent with TEM and FESEM morphological observations, demonstrating that solvothermal treatment enhances graphene sheet exfoliation and can effectively modify graphene-like material structures for more optimal energy storage applications.

Fig. 6 presents the multi-point BET isotherm profile of the synthesized samples, demonstrating a linear correlation between adsorbed gas volume and relative pressure, characterized by a defined slope (s) and intercept (i). These variables make it possible to determine the graphene-like material's monolayer adsorption capacity (V_m) . The BET analysis was performed using a Surface Area Analyzer (SAA) to determine key properties, including specific surface area (m2/g), pore diameter, pore volume, and adsorption isotherm behavior, summarized in Table 1. A higher specific surface area indicates enhanced surface interactions, directly influencing adsorption efficiency. This parameter quantifies the accessible surface for nitrogen (N2) adsorption, providing insights into mass transport within the porous network and potential selectivity in catalytic applications. The adsorption isotherm was recorded over a relative pressure range (P/P_0) of 0.05 to 0.3, following the BET model established in previous studies [5].

The results (Table 1) demonstrate that variations in solvothermal dispersion concentration significantly influence the specific surface area of the samples, suggesting that fine-tuning these parameters can optimize the properties of graphene-like materials. This optimization is advantageous for developing electrodes in electrochemical energy storage systems. Sample SL-2 shows an increase in surface area due to the given dispersion concentration's ability to optimize the reaction conditions between ethylene glycol solvent and precursor, resulting in better porosity than sample SL-1. The optimal sample, SL-2, exhibits the highest specific surface area, enhancing its suitability for electrode applications in energy storage systems. The findings are supported by XRD, FESEM, EDS, and TEM analyses, revealing that SL-2 consists of thin carbon layers, reduced oxygen-containing groups, elevated carbon content, and well-defined sheet-like morphology.

The pore size distribution of the sample produced by nitrogen adsorption at 77.35 K is shown in Fig. 7. Most pores possess diameters between 0 and 10 nanometers. According to the International Union of Pure and Applied Chemistry (IUPAC) standards, pores with diameters ranging from 2 to 50 nanometers are classified as mesopores, confirming the mesoporous nature of the synthesized graphene-like material. The pore size in materials is crucial and beneficial for various practical applications, particularly in fabricating eco-friendly supercapacitor electrodes. The ion transfer rate is a vital determinant for the effectiveness of supercapacitor electrode applications, and high-performance materials exhibiting mesoporous properties have been demonstrated to facilitate rapid and efficient ion transfer. Therefore, mesoporous graphene-like samples with substantial specific surface areas are promising candidates for supercapacitor electrode applications.

The electrochemical charge storage behavior of graphene-like material with solvothermal treatment at the optimal ethylene glycol dispersion concentration (SL-2) and the sample without solvothermal treatment (SL-0) has been analyzed using cyclic voltammetry (CV). Fig. 8 presents the CV profiles obtained at scan rates ranging from 10 to 100 mV/s. The generated CV profiles exhibit characteristic redox features typical of an Electric Double-Layer Capacitor (EDLC), as reported in the literature.

The cyclic voltammetry (CV) profiles depict the variation in current as a function of the applied potential across a supercapacitor or electrode, enabling an evaluation of the material's energy storage capability. The CV curves for SL-0 and SL-2 display a quasi-rectangular shape at various scan rates, which indicates EDLC behavior. Generally, EDLC supercapacitors with rectangular CV curves demonstrate efficient performance and good energy storage capability.

The specific capacitance values were calculated based on the obtained CV data, and the results show a significant difference between the two samples. The SL-2 sample exhibited a specific capacitance of 31.5 F g⁻¹, while SL-0 reached only 6.32 F g⁻¹. It indicates that the graphene-like material produced with solvothermal treatment has a superior energy storage capacity, making it an efficient and sustainable candidate for supercapacitor electrode applications. The enhancement in charge storage efficiency for the SL-2 sample can be attributed to the improved pore structure and surface area resulting from the solvothermal treatment, which strengthens the formation of electric double layers at the electrode-electrolyte interface.

The GCD characterization results in Fig. 9 show changes between the sample without solvothermal treatment (SL-0) and the sample with solvothermal treatment (SL-2). The SL-0 sample has a maximum charge-discharge duration of 31.9 s at a current of 0.5 mA. In contrast, the SL-2 sample exhibits a maximum charge-discharge duration of 39.9 s at a current of 0.5 mA. It indicates that solvothermal treatment with a concentration of 3 mg/ml can enhance the electrical properties of the electrode material. The charge-discharge duration decreases as the applied current increases because the material stores or releases more charge per unit of time at higher currents. The material reaches the cut-off voltage faster at higher than lower currents, resulting in a shorter charge-discharge time.

Fig. 10 presents the Nyquist plot, which displays impedance values in terms of real and imaginary impedance for each sample. The results show that the SL-0 sample has a resistance value of 4.287 Ω , whereas the SL-2 sample exhibits a lower resistance value of approximately 1.87 Ω . Low resistance is a key parameter indicating good electrode quality. Low resistance is essential for supercapacitor electrodes as it enhances energy storage and release efficiency while improving their durability (lifespan). Regarding diffusion impedance, the results show that the SL-0 sample has a diffusion impedance value of 210.8 Ω , while the SL-2 sample exhibits a significantly lower diffusion impedance of around 16.24 Ω . The solvothermal treatment with a concentration of 3 mg/ml applied to the SL-2 sample has been proven to enhance the diffusion capability of the graphene-like material, as indicated by the substantial reduction in diffusion impedance. Lower diffusion impedance facilitates greater ion mobility within the electrode structure. Ions can move more freely and quickly to and from the electrode surface during charge and discharge cycles, improving energy storage capabilities. Enhanced ion transport reduces the time required for charging and discharging, making the supercapacitor more responsive to power demands. The solvothermal treatment with a concentration of 3 mg/ml on the SL-2 sample has been confirmed to improve the electrical properties of the coconut-shell-derived graphene-like material as a supercapacitor electrode.

The material demonstrates significant potential for practical applications. The graphene-like material developed from coconut shell biomass in this study exhibits significant improvements in electrochemical performance, including high specific surface area, low internal resistance, and enhanced specific capacitance. These characteristics make it a promising candidate for electrode applications in energy storage devices, particularly supercapacitors. In the transportation sector, such materials can be utilized in energy storage systems for electric vehicles, which require fast and efficient charge-discharge capabilities. In the telecommunications field and portable electronic devices, this material has potential use in backup power systems and fast-charging technologies. Moreover, utilizing biomass waste such as coconut shell as a raw material supports the principles of sustainability and the circular economy, making this approach not only technically efficient but also environmentally friendly and feasible for industrial-scale implementation.

Despite the promising outcomes of the solvothermal method in enhancing the structure and electrochemical performance of coconut-shell-derived graphene-like materials, this study has several limitations and drawbacks. The reproducibility of the results is sensitive to slight variations in processing parameters, while the material's long-term stabil-

ity and full-cell performance remain unverified. Key disadvantages include the absence of advanced characterization, such as Raman spectroscopy or XPS, which limits deeper structural insights. Moreover, the electrochemical analysis is restricted to CV/GCD/EIS without long-term stability tests. The study is also confined to a single biomass type and solvent, potentially reducing its generalizability.

Future work should explore other biomass sources and green solvents, integrate comprehensive structural and electrochemical analyses, and validate the scalability of the process. Further development may encounter challenges in maintaining material uniformity, balancing porosity and conductivity, and modeling ion transport behavior. However, addressing these issues will be key to unlocking the full potential of biomass-derived graphene-like materials in practical energy storage applications.

7. Conclusions

- 1. Graphene-like materials were successfully synthesized from coconut shell biomass through solvothermal treatment using ethylene glycol as a dispersion medium. The variation in dispersion concentration affected the structural and morphological properties, as indicated by reduced interlayer spacing, the presence of thin nanosheets, increased specific surface area (up to $872.886 \, \text{m}^2/\text{g}$), and decreased intensity of oxygen-containing functional groups.
- 2. Electrochemical analysis revealed that the sample treated with a 3 mg/ml dispersion concentration (SL-2) exhibited enhanced performance, achieving a specific capacitance of 31.50 F/g, reduced internal resistance (1.87 Ω), and extended charge–discharge time. These improvements are attributed to synergistic effects from structural modifications and increased electrochemically active surface area, indicating the potential of dispersion-controlled solvothermal treatment for supercapacitor applications.

Conflict of interest

The authors declare that they have no conflict of interest in relation to this study, whether financial, personal, authorship, or otherwise, that could affect the study and its results presented in this paper.

Financing

This study was funded by the RIIM LPDP Grant and BRIN, grant number (199/IV/KS/11/2023).

Data availability

Manuscript has no associated data.

Use of artificial intelligence

The authors confirm that artificial intelligence tools were utilized within acceptable ethical and scientific boundaries to support this research. All outputs generated by AI have been critically assessed and validated by the authors.

References

- Ahmad, F., Zahid, M., Jamil, H., Khan, M. A., Atiq, S., Bibi, M. et al. (2023). Advances in graphene-based electrode materials for high-performance supercapacitors: A review. Journal of Energy Storage, 72, 108731. https://doi.org/10.1016/j.est.2023.108731
- 2. Mishra, S., Srivastava, R., Muhammad, A., Amit, A., Chiavazzo, E., Fasano, M., Asinari, P. (2023). The impact of physicochemical features of carbon electrodes on the capacitive performance of supercapacitors: a machine learning approach. Scientific Reports, 13 (1). https://doi.org/10.1038/s41598-023-33524-1
- 3. Aimon, A. H., Rahmawati, D., Sutarto, R., Marsudi, M. A., Wibowo, A., Iskandar, F. (2024). Simple and Harmless Fabrication of Reduced Graphene Oxide-Based Transparent Conductive Film Using L-Ascorbic Acid as Reducing Agent. Arabian Journal for Science and Engineering, 49 (7), 10181–10191. https://doi.org/10.1007/s13369-024-09045-y
- $4. \qquad \text{Pei, S., Cheng, H.-M. (2012). The reduction of graphene oxide. Carbon, } 50 (9), 3210-3228. \\ \text{https://doi.org/10.1016/j.carbon.2011.11.010}$
- 5. Widanarto, W., Wulandari, R., Rahmawati, D., Cahyanto, W. T., Sari, K., Effendi, M. et al. (2024). Microwave irradiation-induced yield enhancement of coconut shell biomass-derived graphene-like material. Physica Scripta, 99 (6), 065949. https://doi.org/10.1088/1402-4896/ad4691
- Rahman, M. O., Nor, N. B. M., Sawaran Singh, N. S., Sikiru, S., Dennis, J. O., Shukur, M. F. bin Abd. et al. (2023). One-Step Solvothermal Synthesis by Ethylene Glycol to Produce N-rGO for Supercapacitor Applications. Nanomaterials, 13 (4), 666. https://doi.org/10.3390/ panol 3040666
- 7. Pham, P. V., Mai, T.-H., Dash, S. P., Biju, V., Chueh, Y.-L., Jariwala, D., Tung, V. (2024). Transfer of 2D Films: From Imperfection to Perfection. ACS Nano, 18 (23), 14841–14876. https://doi.org/10.1021/acsnano.4c00590
- 8. Novoselov, K. S., Geim, A. K., Morozov, S. V., Jiang, D., Zhang, Y., Dubonos, S. V. et al. (2004). Electric Field Effect in Atomically Thin Carbon Films. Science, 306 (5696), 666–669. https://doi.org/10.1126/science.1102896
- Dreyer, D. R., Park, S., Bielawski, C. W., Ruoff, R. S. (2010). The chemistry of graphene oxide. Chem. Soc. Rev., 39 (1), 228–240. https://doi.org/10.1039/b917103g
- 10. Wang, Y., Duan, Y., Liang, X., Tang, L., Sun, L., Wang, R. et al. (2023). Hierarchical Porous Activated Carbon Derived from Coconut Shell for Ultrahigh-Performance Supercapacitors. Molecules, 28 (20), 7187. https://doi.org/10.3390/molecules28207187
- 11. Widanarto, W., Solehudin, H., Rahmawati, D., Byennardsi, S. D., Effendi, M., Ghoshal, S. K., Kurniawan, C. (2025). Lanthanum-doped rGO from biomass: a sustainable electrode material for enhanced supercapacitor performance. Engineering Research Express, 7 (2), 025004. https://doi.org/10.1088/2631-8695/adca8b
- 12. Huo, Y., Xiu, S., Meng, L.-Y., Quan, B. (2023). Solvothermal synthesis and applications of micro/nano carbons: A review. Chemical Engineering Journal, 451, 138572. https://doi.org/10.1016/j.cej.2022.138572
- 13. Mahmun, A., Deoghare, A. B. (2024). A comparative study on coconut shell-derived graphene oxide and reduced graphene oxide. Current Applied Physics, 62, 12–21. https://doi.org/10.1016/j.cap.2024.03.009
- Gupta, B., Kumar, N., Panda, K., Kanan, V., Joshi, S., Visoly-Fisher, I. (2017). Role of oxygen functional groups in reduced graphene oxide for lubrication. Scientific Reports, 7 (1). https://doi.org/10.1038/srep45030
- 15. Shams, M., Guiney, L. M., Huang, L., Ramesh, M., Yang, X., Hersam, M. C., Chowdhury, I. (2019). Influence of functional groups on the degradation of graphene oxide nanomaterials. Environmental Science: Nano, 6 (7), 2203–2214. https://doi.org/10.1039/c9en00355j
- 16. Carrera, C., Galán-González, A., Maser, W. K., Benito, A. M. (2025). Multifaceted role of H2O2 in the solvothermal synthesis of greenemitting nitrogen-doped graphene quantum dots. Chemical Science, 16 (8), 3662–3670. https://doi.org/10.1039/d4sc07896a
- 17. Popov, I. A., Bozhenko, K. V., Boldyrev, A. I. (2011). Is graphene aromatic? Nano Research, 5 (2), 117–123. https://doi.org/10.1007/s12274-011-0192-z
- 18. Yu, W., Sisi, L., Haiyan, Y., Jie, L. (2020). Progress in the functional modification of graphene/graphene oxide: a review. RSC Advances, 10 (26), 15328–15345. https://doi.org/10.1039/d0ra01068e
- 19. Nethravathi, C., Rajamathi, M. (2008). Chemically modified graphene sheets produced by the solvothermal reduction of colloidal dispersions of graphite oxide. Carbon, 46 (14), 1994–1998. https://doi.org/10.1016/j.carbon.2008.08.013
- 20. Sankar, S., Lee, H., Jung, H., Kim, A., Ahmed, A. T. A., Inamdar, A. I. et al. (2017). Ultrathin graphene nanosheets derived from rice husks for sustainable supercapacitor electrodes. New Journal of Chemistry, 41 (22), 13792–13797. https://doi.org/10.1039/c7nj03136j