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Utilization of Ultrasonic Wave in the Production of Reduced Graphene Oxide from Coconut Shell Biomass: Eco-Friendly and sustainable Approach

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Abstract

The production of reduced graphene oxide (rGO) using environmentally friendly methods remains a challenge in the development of sustainable energy storage materials. This study explores the utilization of ultrasonic waves in the production of rGO from coconut shell biomass as a green and cost-effective approach. Ultrasonic treatment for 30 minutes (UB-30) resulted in a graphene sheet morphology, enhanced carbon content, and reduced oxygen functional groups on rGO. Electrochemical characterization showed that the specific capacitance of the ultrasonically treated rGO (UB-30) reached 789 F/g at a scan rate of 10 mV/s, demonstrating competitive electrochemical performance for supercapacitor applications. The use of coconut shell biomass as a precursor offers an eco-friendly solution, while the application of ultrasonication enables higher production efficiency with lower energy consumption. These findings contribute significantly to the development of electrode materials for supercapacitors and sustainable energy storage systems.

Introduction

Graphene is a single layer of carbon atoms organized in a two-dimensional honeycomb structure, displaying outstanding mechanical, thermal, and electrical characteristics. It is renowned for its extraordinary strength, excellent electrical conductivity, and extensive surface area, which make it ideal for use in applications such as electronics, energy storage, and composite materials [1-4]. Various modifications have been developed, including the addition of doping, structural engineering, and chemical functionalization. One of the most prominent modifications is graphene oxide (GO), which is through the oxidation of graphene to obtain oxygen functional groups that can increase its reactivity and compatibility in various applications. Graphene Oxide (GO) is a modified form of graphene that features a variety of oxygen-containing functional groups, including hydroxyl, carboxyl, and epoxide groups. These functional groups give GO hydrophilic properties, which improve its ability to disperse in water-based solutions. However, their presence interrupts the π-conjugation system found in graphene, leading to a substantial reduction in electrical conductivity compared to pure graphene [5,6].

Graphene oxide (GO) with abundant oxygen functional groups offers high flexibility in nanomaterial-based applications. Nanomaterials have nanometer-scale structures and offer unique properties such as large surface area, unique optical properties, and tunable conductivity, making them widely used in energy, catalysis, and biomedical technologies. Nanomaterials based on graphene and graphene oxide have attracted widespread research interest due to their distinct physicochemical properties [7]. However, for needs that require better electrical conductivity or material properties approaching pure graphene, further development can be done through reduced graphene oxide (rGO). This process produces materials with improved electronic characteristics, making it one of the promising nanomaterials in energy technology, sensors, and electronic devices. Reduced Graphene Oxide (rGO) is created by chemically or thermally reducing Graphene Oxide (GO) to eliminate some of the oxygen-containing functional groups. This reduction raises the carbon-to-oxygen (C/O) ratio, resulting in improved electrical conductivity (up to 6300 S cm^{-1}) and mechanical characteristics that more closely resemble those of pure graphene [8,9].

Natural graphite has been selected as the raw material for producing rGO with high potential [10]. One of the natural materials commonly used as a carbon source is coconut shell, which offers a sustainable and abundant supply of biomass for various applications, such as the production of carbon-based nanomaterials like graphene oxide. The conversion of coconut shell waste into graphene oxide (GO) and reduced graphene oxide (rGO) represents a promising and sustainable approach to graphene production. Coconut shells have a high carbon content (ranging from about 49.86% to 74.3%), which makes them a suitable biomass for producing graphene-based materials [11]. Utilizing coconut shell waste not only helps to minimize environmental impact but also supports sustainability within the agricultural industry. Coconut shells have a high carbon content, with research showing that charcoal made from them can yield up to 57.11% carbon. The use of coconut shell waste for graphene production not only helps solve waste management problems but also offers economic benefits to the agricultural sector by generating value-added products [12]. The physical characteristics of rGO are highly influenced by the spatial arrangement of functional groups, the shape and size of the particles, as well as structural defects. Thus, gaining a deeper understanding of how the GO structure evolves during the reduction and exfoliation processes is crucial for achieving the desired physical properties of rGO [13] .

Ultrasonic methods have gained recognition as an effective strategy for synthesizing reduced graphene oxide (rGO) from graphene oxide (GO) and other starting materials. These techniques utilize the physical effects of ultrasound, including cavitation and shear forces, to enhance the reduction and exfoliation processes necessary for producing rGO. The ultrasonication process results in thinner graphene oxide layers [14]. Generally, graphene is produced from graphite as a carbon source. Various methods for synthesizing graphene from graphite as the base material have been developed, with the reduced graphene oxide (rGO) method being one of the most commonly used. This process starts with synthesizing graphite into graphite oxide, followed by the oxidation of graphite into graphene oxide (GO) using a strong oxidizing agent, and finally, the oxide bonds in GO are reduced using a reductant [15,16]. The rGO method offers the advantage of producing graphene in large quantities, but it involves multiple complex steps, various chemicals, and an extended synthesis process. In this study, a modified approach to the reduced graphene oxide (rGO) method was adopted by simplifying some synthesis steps and substituting graphite with activated carbon derived from coconut shell waste as the carbon source. Using coconut shells to produce activated carbon presents a promising alternative for graphene synthesis. This method provides several benefits, including simpler synthesis steps, easier processing, and the potential for large-scale production. The objective of this research is to utilize coconut shell waste to create activated carbon as the starting material for graphene synthesis through the modified rGO method using ultrasonic.

Experimental Method

The research was divided into several key stages: carbonization, activation, exfoliation, and reduction, as depicted in Figure 1. Coconut shells, the primary raw material, were sourced from Banyumas Regency. After being cleaned and dried, the shells were carbonized at 300 °C for 1 hour to produce charcoal, which is hereafter referred to as carbon. The carbon was then activated using potassium hydroxide (KOH) at a mass ratio of 1:4 (carbon to KOH). The activation process lasted for 24 hours, followed by repeated washing with deionized water until a neutral pH was achieved, ensuring the removal of residual KOH and other impurities. The activated carbon was subsequently exfoliated to produce graphene oxide (GO) via ultrasonication at a frequency of 40 kHz and a power of 120 watts. To examine the effect of sonication duration on GO quality, different ultrasonication times were employed: 30 and 60 minutes, designated as UB-30 (30 minutes of ultrasonication) and UB-60 (60 minutes of ultrasonication), respectively. As a control, a sample without ultrasonication treatment was prepared, labeled UB-0. After exfoliation, GO was reduced using a microwave at 800 watts for 5 minutes to eliminate oxygen-containing functional groups. The reduced graphene oxide (rGO) was then dried at 100 °C for 24 hours to yield rGO powder. The final material was characterized using a range of techniques, including FE-SEM, EDS, XRD, and electrochemical testing, to assess the morphology, elemental composition, crystal structure, and electrochemical properties of the synthesized rGO.

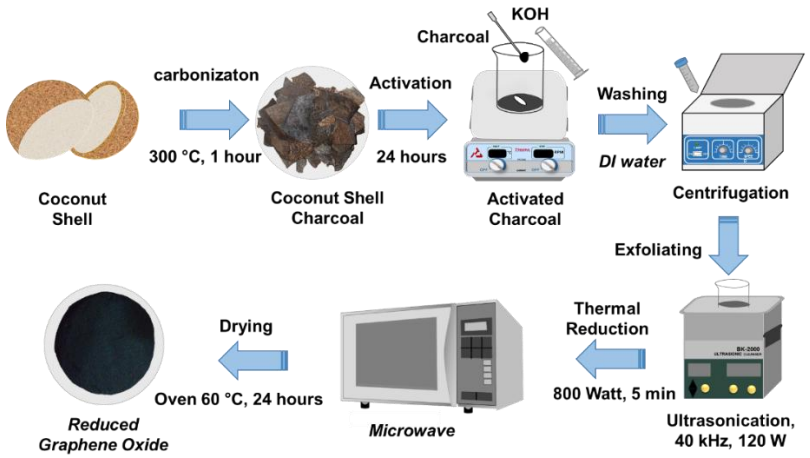


Figure 1. Synthesis of reduced graphene oxide from coconut shell.

The preparation for electrochemical testing involved synthesizing electrode materials with different ultrasonic treatment durations (UB-0, UB-30, UB-60). The active material was then mixed uniformly with a conductive agent and a binder in a specific ratio to form a slurry, which was applied to a current collector. After drying to remove solvent residues, the electrodes were pressed to ensure uniformity and assembled with a suitable electrolyte for cyclic voltammetry (CV) analysis under consistent testing conditions.

Result and Discussion

The rGO produced without ultrasonic treatment (UB-0) and the rGO treated with ultrasonic waves for 30 and 60 minutes (UB-30 and UB-60) exhibit diffraction patterns as shown in Figure 2.

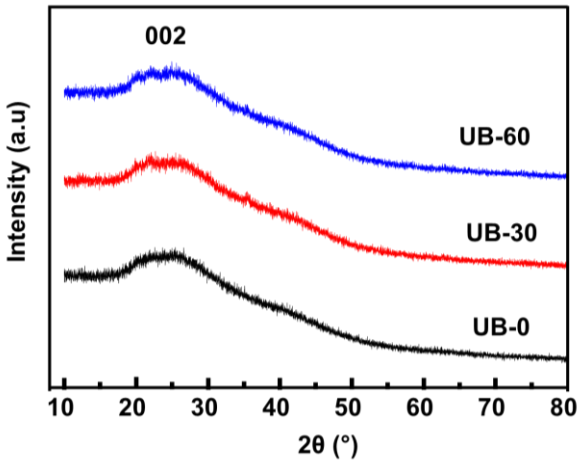
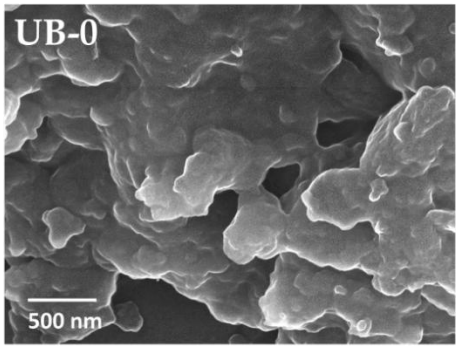
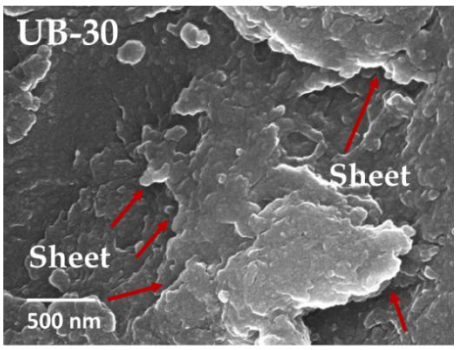


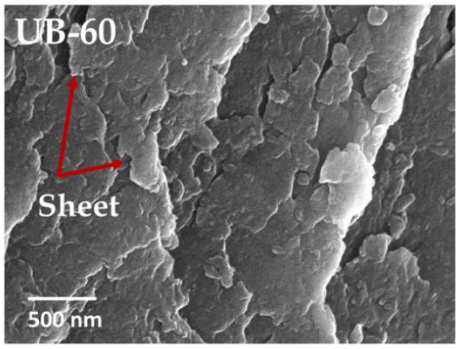
Figure 2. XRD Analysis of Reduced Graphene Oxide varying ultrasonication durations (0, 30, and 60 minutes).

Figure 2 shows the diffraction pattern of the produced samples. The diffraction pattern exhibits a broad dominant peak at 2θ between 20° and 30°, corresponding to the (002) plane orientation of the graphite lattice. This XRD pattern indicates disruptions between carbon

layers and the formation of graphene within the porous carbon structure [17-19]. In the samples treated with ultrasonic waves (UB-30 and UB-60), the diffraction peak slightly broadens and decreases in intensity. This suggests an increased interlayer spacing due to enhanced exfoliation, as well as a reduction in oxygen groups through the removal of oxide groups. Although the difference is not significant, the peak broadening also indicates an increase in structural disorder, likely caused by defect formation during the exfoliation process. These results support the hypothesis that ultrasonic treatment plays a role in modifying the internal structure of carbon and has the potential to enhance the characteristics of rGO materials.







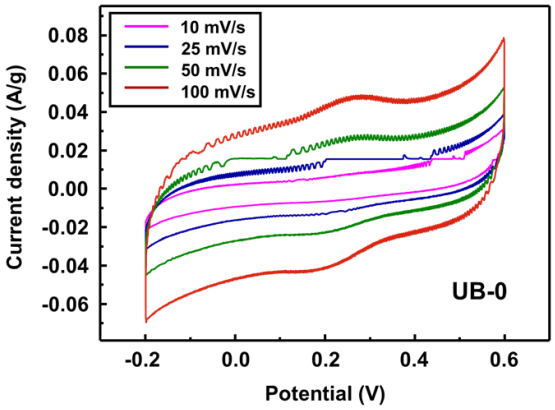
n	UB-0	UB-30	UB-60
C	47.9	52.1	46.0
O	47.5	42.0	46.0
K	4.6	5.9	8.0

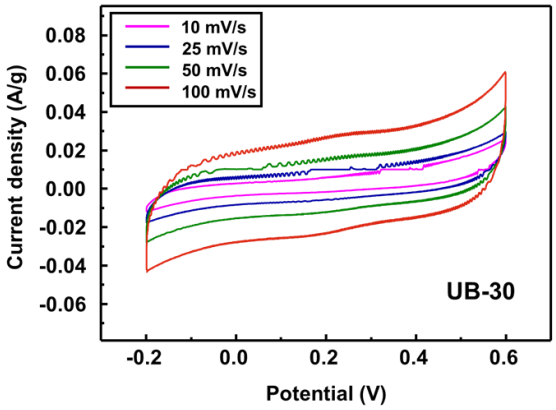
n = element quantification

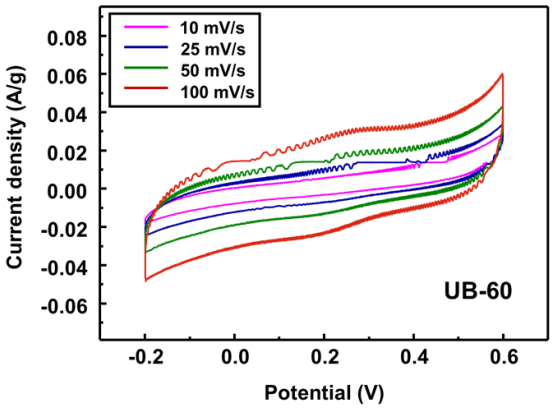
Figure 3. FE-SEM images and quantitative elemental analysis of reduced graphene oxide samples with varying ultrasonication durations (0, 30, and 60 minutes).

Figure 3 shows the results of a Field Emission-Scanning Electron Microscope (FE-SEM) with a magnification of 150,000x. This magnification allows detailed observation of pores, cracks, and particle aggregation, which are important indicators in understanding material properties, such as specific surface area, mechanical properties, and the ability to interact with the environment. Figure 3 presents the FE-SEM micrographs and quantitative elemental analysis of the samples produced without and with ultrasonic treatment at various durations. The UB-0 sample exhibits the formation of dense agglomerates with uneven distribution. In contrast, ultrasonic treatment in the UB-30 and UB-60 samples resulted in more distinct exfoliated structures, with longer ultrasonic durations promoting the formation of thin sheets resembling graphene. This morphology aligns with the graphene characteristics reported in the literature[20] . The EDX analysis confirms the corresponding atomic composition, where the UB-30 sample has the highest carbon (C) content and was selected as the optimal sample for

further electrochemical performance testing. These findings indicate that ultrasonic treatment effectively reduces oxygen (O) functional groups and increases carbon (C) content in the material. This is consistent with previous studies, which have reported that ultrasonication can exfoliate materials into thin sheets by reducing oxygen groups, thereby enhancing carbon content [21], [22].







Scan Rate (mV/s)	Specific Capacitance (F/g)		
	UB-0	UB-30	UB-60
10	1231	789	875
25	928	580	608
50	805	521	509
100	722	462	438

Figure 4. CV curve and specific capacitance of reduced graphene oxide samples with varying ultrasonication durations (0, 30, and 60 minutes).

Figure 4 presents the CV curves and specific capacitance of the supercapacitor electrodes from the UB-0, UB-30, and UB-60 samples. The cyclic voltammetry (CV) analysis of the three samples demonstrates charge storage characteristics resembling those of electric double-layer capacitors (EDLC), consistent with previous reports in the literature [23-27]. In all three samples, an increase in the scan rate leads to a decrease in capacitance values. This phenomenon is attributed to ion diffusion limitations in the electrolyte. At lower scan rates, the electrolyte ions have sufficient time to diffuse into the material's pores, maximizing charge storage. However, at higher scan rates, the charging and discharging processes occur more rapidly, limiting ion access to the electrode surface and macro-pores. The micro-pores, which contribute more significantly to capacitance, are not fully utilized, resulting in reduced capacitance values.

Interestingly, the UB-0 sample, which did not undergo ultrasonic treatment, exhibited relatively high capacitance at various scan rates. This could be due to the simpler amorphous

carbon structure of UB-0, allowing easier diffusion of electrolyte ions throughout the material's pores, even at higher scan rates. Additionally, the presence of more oxygen functional groups in UB-0 may contribute to stronger electrostatic interactions with the electrolyte ions. These oxygen groups help create a more stable electrostatic double layer around the electrode surface, promoting more efficient ion distribution into the material's pores and increasing charge storage. The UB-30 sample, subjected to 30 minutes of ultrasonic treatment, had a lower specific capacitance compared to UB-0 and UB-60. This could be attributed to the restacking phenomenon of the carbon sheets after exfoliation, where the sheets reassemble. This restacking obstructs the diffusion of electrolyte ions across the entire active surface, limiting ion access and thus reducing the specific capacitance. In the UB-60 sample, which underwent ultrasonic treatment for 60 minutes, an increase in specific capacitance was observed. Longer ultrasonic treatment further opens the carbon structure, improving electrolyte ion accessibility to the entire electrode surface, contributing to better charge storage efficiency.

Conclusion

This study successfully demonstrates the utilization of ultrasonic waves in the production of reduced graphene oxide (rGO) from coconut shell biomass with an eco-friendly and sustainable approach. Ultrasonic treatment for 30 minutes resulted in a graphene sheet morphology, enhanced carbon content, and reduced oxygen functional groups on rGO. The specific capacitance value of UB-30 at a scan rate of 10 mV/s reached 789 F/g, demonstrating competitive electrochemical performance for supercapacitor applications. The use of coconut shell biomass as a precursor offers an environmentally friendly alternative, while the application of ultrasonication provides an efficient and cost-effective rGO production process. These findings significantly contribute to the development of sustainable energy storage materials and open up broader application opportunities in supercapacitors and other energy storage devices.

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