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Synthesis and Characterization of Electrochemically Reduced Graphene Oxide for Applications in Supercapacitors

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ABSTRACT

Graphene oxide (GO) was prepared from graphite by the modified Hummers' method using strong oxidizing agents which was reduced removing its oxygen-containing functional groups. sp^2 carbon domains in GO material do not allow for electron transferring due to tight geometry and higher electronegativity of sp^2 orbitals than sp^3 carbon centers. Successful synthesis of GO from graphite powder was confirmed by X-ray diffraction (XRD) technique showing a diffraction peak $2\theta = \sim 9.8^\circ$. Further, GO reduction procedures were performed through electrochemical (three-electrode cell, 1 M KOH, potential range from 0 to -1.2 V) and chemical (1 M ascorbic acid, 60 °C, stirring) methods. Morphological (scanning electron microscopy (SEM) and transmission electron microscopy (TEM)), structural (X-ray photoelectron spectroscopy (XPS) and Raman spectroscopy) and thermal (thermogravimetric analysis (TGA)) analyses confirmed the removal of oxygen-containing groups, indicating conversion from GO to reduced GO (rGO). The electrochemical characteristics (CV, GCD, EIS) were investigated in a symmetric two-electrode Swagelok-type cell using 1 M NaNO_3 . Based on the obtained results, the most promising material was electrochemically reduced graphene oxide (E-RGO), where the capacitance increased from $\sim 54 \text{ F} \cdot \text{g}^{-1}$ at 0.7 V to $\sim 67 \text{ F} \cdot \text{g}^{-1}$ at 1.5 V at a specific current density of $200 \text{ mA} \cdot \text{g}^{-1}$ in a 1 M NaNO_3 electrolyte. The specific capacitance of the E-RGO electrodes is 13.7% higher than that of chemically reduced graphene oxide (C-RGO), and these results demonstrate the effectiveness of electrochemically reduced graphene oxide, further demonstrating its potential applications in supercapacitors.

1. Introduction

The use of regenerative materials can address significant challenges associated with current issues regarding energy storage in supercapacitors. Supercapacitors offer fast charging and discharging rates, high power density, and excellent cycling stability. Despite these distinctive features, supercapacitors have the following limitations in modern applications. First, the accumulation of charge by electrostatic means through ion adsorption results in low energy densities for traditional double-layer super-

capacitors (EDLCs). Compared to lithium-ion batteries, they have energy densities ranging from 5 to $15 \text{ W} \cdot \text{kg}^{-1}$ [1]. Second, strong van der Waals forces act in pure graphene and graphene oxide, reducing the area of the electrochemically active surface by clustering individual nanolayers. These nanolayers significantly hinder the penetration of electrolyte ions during ion transport [2]. Third, contact between the electrode and the electrolyte is limited due to graphene's low wettability. It is precisely for this reason that ions reach the internal pores slowly. Thus, most of the material's active surface re-

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mains unaffected during ion transport [3]. It should be emphasized that the complete removal of oxygen-containing functional groups by chemical or thermal means can lead to the loss of pseudocapacitive redox centers. These centers provide additional charge storage [4]. To avoid electrochemical and structural limitations, electrodes based on graphene oxide-derived reducing materials must meet specific criteria for long-term cycling stability and operating potential windows [5]. The porous structures of rGO exhibit a specific capacitance ranging from 150 to 475 F·g⁻¹, depending on the electrolyte and the synthesis method [6]. Most electrochemical reduction reactions use aqueous electrolytes, and in these cases, the electrolytes operate within a voltage range of 1.0–1.2 V. This prevents water decomposition above 1.3–1.5 V. To extend the potential range to 2.5–3.0 V, it is recommended to use organic liquids. An important requirement when working with such liquids is to reduce the number of electrochemically active oxygen-containing functional groups. Otherwise, side reactions may occur at high anode potentials. For the practical application of rGO electrodes in supercapacitors, long-term electrochemical durability is required during repeated charge-discharge cycles. According to A.Z. Abdulahi, rGO electrodes retain 90–97% of their capacitance even after 10,000 cycles [3].

The history of GO extends back centuries, with the first well-known example of graphite oxidation achieved by B. Brodie in 1859 using potassium chlorate and nitric acid (typically about 90–100% HNO₃). The presence of oxygenated functional groups introduce strong hydrophilicity to oxidized graphene, enabling its stable dispersion in distilled water and other organic media. This enhanced dispersibility is particularly advantageous for the fabrication of thin films and composite materials. Furthermore, the oxidation process leads to a significant expansion of the interlayer spacing, increasing from approximately 0.334 nm in pristine graphite to about 0.6–1.2 nm in GO, depending on the extent of water molecule intercalation [7].

The most common and widely cited method currently used for GO synthesis is the Hummers' method (using KMnO₄, NaNO₃, and H₂SO₄) or modified versions thereof, such as those omitting NaNO₃ and utilizing a mixture of H₂SO₄/H₃PO₄ for improved efficiency and safety [8]. The Hummers' method is a common technique for producing graphene oxide from graphite. In this process, graphite is oxidized in a mixture of strong acids and potassium permanganate. The layers are then exfoliated to produce a

colloidal suspension of GO. This oxidation process introduces many oxygenated functional groups, such as epoxy, carboxyl, and hydroxyl groups. In result, up to ~55% of carbon atoms form C–O bonds [9]. The ability to reduce GO – restoring the sp² network and recovering electrical conductivity is crucial for its widespread application in energy storage and electronic devices. The residual oxygen defect density can be tailored for covalent functionalization, enabling selective detection of specific chemical or biological targets [10].

GO is a key material with broad applications due to its versatile structure. GO is obtained from oxidized graphite synthesized using the modified Hummers' method and naturally exists in both monolayer and multilayer forms. In the modified Hummers' synthesis (during the oxidation process), oxygen-containing functional groups are introduced at the edges of the carbon lattice. The availability of GO as a scalable and processable intermediate material makes its reduction one of the most important reactions in graphene chemistry [11]. The introduction of oxygen-containing functional groups significantly disturbs the original sp²-bonded carbon network present in pristine graphite. As a result, graphene oxide exhibits distinct properties, including electrical insulation, increased hydrophilicity, improved dispersibility in solvents, and expanded interlayer spacing. In contrast to graphene, GO behaves as an electrical insulator because the continuous sp² network is interrupted by sp³-hybridized carbon atoms [12]. Common oxygen-containing functional groups include hydroxyl (–OH), epoxy (–O–C–), carboxyl (–COOH), and carbonyl (–C=O) groups. The carboxyl (–COOH) and carbonyl (–C=O) groups are located at the edges of the sheet, while others: hydroxyl (–OH) and epoxy (–O–C–) are located in the basal plane [6]. These oxygen-containing functional groups ensure high dispersibility in water and other solvents. However, these properties reduce its charge transport capability compared to pure graphene [13].

E-RGO is also one of the important graphene-based materials. Electrochemical reduction (ER) is becoming increasingly popular compared to traditional reduction methods [14]. This process removes oxygen-containing groups from graphene oxide (Fig. 1). For example, chemical reduction methods often involve the use of hazardous and toxic substances such as hydrazine, which can leave behind undesirable residues. Thermal reduction processes, on the other hand, can damage the graphene structure due to high temperatures (800 °C) [15, 16].

In contrast, ER offers a cleaner and more environ-

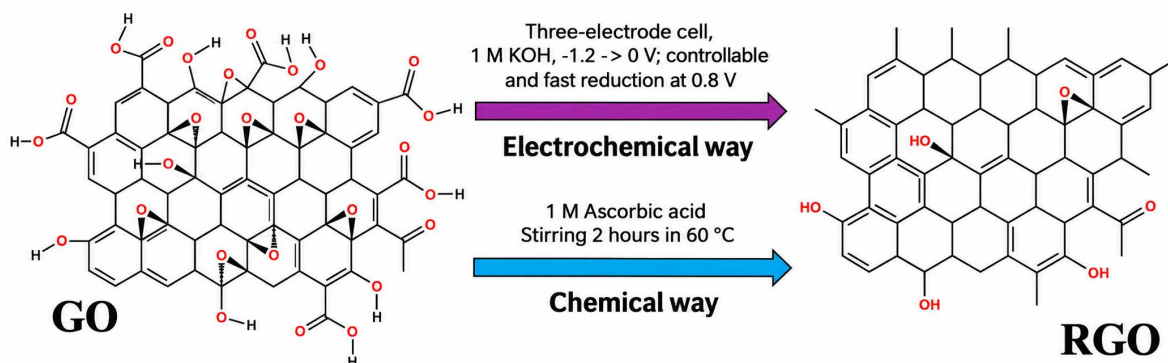


Fig. 1. Schematic illustration of graphene oxide layer (GO) losing its oxygen-containing functional groups to form reduced graphene oxide.

mentally friendly alternative. Another advantage of ER is its green methods and efficiency. The process is relatively fast, cost-effective, and easy to control by adjusting parameters such as the applied potential and reduction time. Typically, it is carried out in a three-electrode setup using conductive substrates like stainless steel foil (SS 316L) [17]. As oxygen groups are removed, the carbon-to-oxygen (C/O) ratio rises significantly, leading to much better electrical conductivity. The resulting E-RGO is stable and highly applicable for various applications, including energy saving devices like supercapacitors, as well as biosensors and electrocatalytic systems [18].

Electrochemical reduction can be carried out by direct reduction in an organic gel suspension (a one-step process) or by first depositing the material followed by reduction (a two-step process). E-RGO is obtained by cycling a graphene oxide film between $0 \rightarrow -1.5$ V under N_2 atmosphere 0.1 M Na_2SO_4 , typically 22 cathodic scans, which partially removes oxygen groups while preserving a conductive sp^2 network [19]. Electrochemical reduction is one of several routes (chemical, thermal, and electrochemical) to restore the conductive graphene lattice. The resulting electrochemically reduced graphene oxide retains a high specific surface area and a tunable electronic structure, offering both improved conductivity and residual functional groups for hybridization with electrocatalysts [20]. Chemical Reduction is a scalable method often utilizing strong reducing agents such as hydrazine, $NaBH_4$, or hydroiodic acid (HI). While effective, this route commonly introduces toxic or hazardous chemicals (e.g., hydrazine) or leaves behind undesirable residues, which can compromise the product quality and pose environmental issues [21]. Recent advances in chemical reduction focus on utilizing environmentally friendly (green) reducing agents, like gallic acid or ascorbic acid (AA),

to address toxicity concerns [22–25]. In our proposed research work, C-RGO) was synthesized using ascorbic acid as the reducing agent. Thermal Reduction involves rapid heat treatment (often above $1000^\circ C$) to remove oxygen functionalities. However, the extreme temperatures often lead to the formation of structural defects such as vacancies due to the rapid release of gases (CO_2 and CO). Research is now focusing on low-temperature thermal annealing ($300^\circ C$) to achieve suitable conductivities while maintaining substrate compatibility for flexible electronics [26]. When it comes to electrochemical type of reduction of graphene oxide is a comfortable and safe method for producing reduced material, as it uses aqueous electrolytes at a room temperature. This helps avoid the involvement of toxic substances and high thermal conditions [27].

In this scientific work, graphite powders were oxidized in highly concentrated ($>96\%$) sulfuric acid (H_2SO_4) and hydrogen peroxide (H_2O_2) solution at fixed temperatures (low-high depended on reaction step). Following exfoliation, GO powders were obtained. In order to remove oxygenated functional groups from single-layered graphene oxide, electrochemical and chemical reduction processes were implemented for comparison purposes.

The main novelty of this work lies in the fact that effective reduction can be achieved through precise control of the electrochemical potential ranges without changing the temperature. The reduction procedure was performed in an alkali solution (1 M KOH) using a three-electrode system in which SS316L stainless steel foil was used as the working electrode. This electrochemical approach helps to increase the reduced material scalability. Furthermore, the specific capacitance of electrodes made from E-RGO exceeds that of C-RGO materials, eliminating the need for chemical reducing agents.

2. Experimental part

2.1. Materials

For the synthesis of graphene oxide (GO), artificial graphite powder (C, AGP-2L-S, anode grade) supplied by Shandong Gelon Lib Co., Ltd. (Linyi, Shandong, China) was used as the carbon precursor. Concentrated sulfuric acid (H_2SO_4 , 93.6–95.6 wt.%, analytical grade) was supplied by Merck KGaA (Darmstadt, Germany). Potassium permanganate (KMnO_4 , analytical grade, China), sodium nitrate (NaNO_3 , chemically pure (CP), Russia), hydrogen peroxide (H_2O_2 , 37 wt.%, Russia), and hydrochloric acid (HCl , 35 wt.%, chemically pure (CP), Russia) were used during the oxidation, termination, and washing steps. Chemical reduction of GO was carried out using L-ascorbic acid ($\text{C}_6\text{H}_8\text{O}_6$, >99.0%) supplied by Shanghai Macklin Biochemical Co., Ltd. (Shanghai, China) as the reducing agent. For electrode preparation, polytetrafluoroethylene (PTFE) dispersion ($(\text{C}_2\text{F}_4)_n$, 60 wt.% in H_2O , Sigma-Aldrich, USA) was used as a binder, while conductive carbon black C65 (C, volatile content $\leq 0.15\%$, China) was used as the conductive additive. 2-propanol (isopropyl alcohol, $\text{C}_3\text{H}_8\text{O}$, HPLC grade, 99.9%, Sigma-Aldrich, France) was used as cleaning reagent. For the electrochemical reduction experiments, potassium hydroxide (KOH , 95%) supplied by Shanghai Macklin Biochemical Co., Ltd. (Shanghai, China) was used to prepare the alkaline electrolyte.

2.2. Graphene oxide synthesis

First an ice bath was prepared and maintained at 0–3 °C. In the bath, a round-bottom flask and a magnetic stirrer were put, after which 1 g of Sodium nitrate salt (NaNO_3) was weighed in the flask and 70 mL of concentrated H_2SO_4 was added. The ice bath was put to stirring mode containing the mixture for 10–15 min to achieve total disintegration. After 10–15 min 2 g of graphite was added into the mixing solution and stirred for 30 min in an ice bath. After 30 min, 8 g of potassium permanganate (KMnO_4) was slowly added to the reaction mixture in small portions while maintaining the temperature in the ice bath. After the addition, the mixture was stirred for an additional 30 min. The solution changes its color from black to black-green after adding KMnO_4 . The temperature was then increased to 35–40 °C by replacing the ice-water bath with a heated antifreeze bath. After that, the reaction mixture was displaced to a vessel maintained at a temperature of 35–40 °C, where it was stirred continuously for 12 h. The next morning, the temperature was in-

creased up to 60–65 °C, after which 240 mL of distilled water was added dropwise. After the water was fully added, the mixture was stirred unstopped for 2 h at the same temperature. During the reaction, the color of the suspension transformed from black-green to black-brown. Then the temperature was raised to 92–98 °C to add 25 mL of a 30% H_2O_2 solution. After adding the hydrogen peroxide, the mixture was homogenized for another 5 min at the same temperature.

In the final stage, the resulting oxidized material mixture was periodically washed in centrifuge (6000 rpm, 10 min) with hydrochloric acid, in order to extract Mn^{2+} ions. Then, purified 6–7 times with deionized water until the pH value indicated a neutral environment. Finally, the obtaining material at the end was dried in a vacuum oven at 60 °C for 12 h (720 min). After drying, a thin GO film was obtained. Using an agate mortar, the film was ground into fine powder particles. In total, 2.95 g of films were obtained, which will then use for electrochemical studies. Schematic diagram representation illustrated in following image (Fig. 2).

2.3. Chemical reduction of graphene oxide

GO is chemically reduced using ascorbic acid. Ascorbic acid is considered a safe and environmentally friendly reducing agent. First, 150 mL of 1 M ascorbic acid solution was prepared in a 250 mL flask. It was then stirred at 60 °C for 2 h. Next, 200 mg of graphene oxide powder was added. Subsequently, the flask was tightly sealed, and the reaction was carried out at ambient room temperature for 24 h. During this process, the color of the particles changed from brown (graphene oxide in fine particles) to black (chemically reduced graphene oxide). This indicates that the material was successfully reduced using ascorbic acid. At the end of the reduction process, the resulting material was washed with distilled water using vacuum filtration. The C-RGO was then dried in a vacuum oven for 24 h at 120 °C. The result was a dry black powder.

2.4. Electrochemical reduction of graphene oxide

After obtaining dark brown graphene oxide powders from graphite using modified Hummers' method, an ER procedure was performed. To do this, we prepared a 5 mg·mL⁻¹ dispersion with distilled water. 5 mg·mL⁻¹ GO dispersion was reduced in a standard three-electrode system using electrochemical methods. The homogeneous dispersion then deposited through drop-casting technique onto the surface of

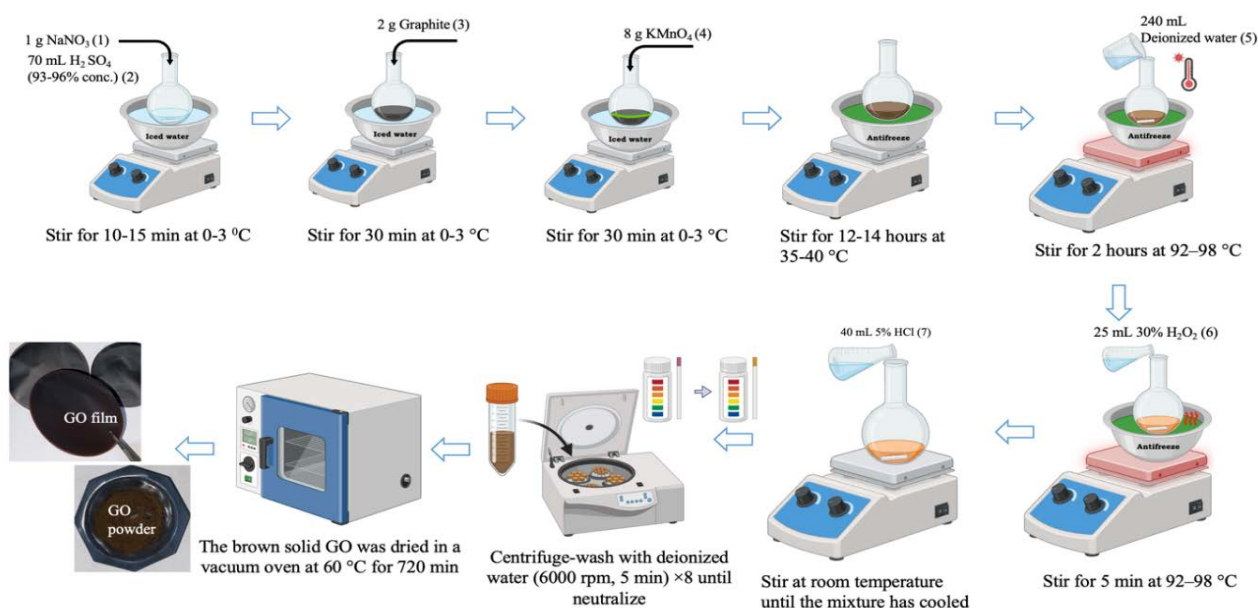


Fig. 2. Schematic representation of step-by-step synthesis process of Gt-O from Graphite by modified Hummer's method.

stainless steel (SS316L) foils, which had been cleaned with ethanol, after that these GO slurry coated substrates were dried in an oven at 60 °C for 12 h to ensure uniform coating. After coating and drying, the SS316L served as the conductive working electrode (WE). As a counter electrode (CE) a Titanium mesh was used and an Ag/AgCl used as reference electrode (RE) (3.5 M KCl). No separator was placed between the working and counter electrodes during the electrochemical reduction experiment. ER was performed using CV with a potentiostat/galvanostat (SP-150e Biologic, France) in the potential range from 0 to -1.2 V, with a scan rate of $5 \text{ mV} \cdot \text{s}^{-1}$ in 6 cycles. The reduction process was investigated in 1 M KOH electrolyte (Fig. 3).

Typically, the ER of GO in aqueous electrolyte is carried out in the voltage window of 0 to -1.5 V. Scan speed in the range of 20 to $100 \text{ mV} \cdot \text{s}^{-1}$ [28, 29]. It has been reported that the CV obtained from the ER of GO from an aqueous suspension at a working electrode exhibits two cathodic peaks [30]. As demonstrated in Fig. 3, the controllable reduction peak widely appeared in -0.8 V and -1.1 V. Guo et al. [31] showed that the C=O and O-H functional groups could be converted between -0.7 to -0.8 with the extension of reduction time. Further, C-O-C group could only be reduced in -1.5 V [28]. However, a more negative potential than -1.5 V could lead to hydrogen bubble evolution. Instead of uniformly reducing the material it can cause defects at the edges of GO structural lattice. Nevertheless, one most common observation in the CV of the GO/SS is that the cathodic current peak

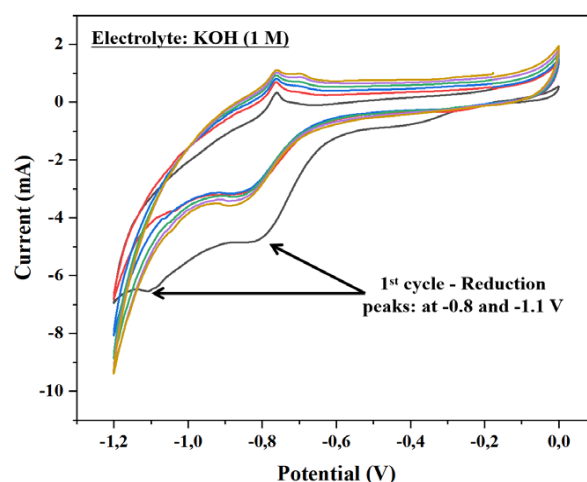


Fig. 3. CV reduction curves of the GO coated SS316L in 1 M KOH over the potential range from 0 to -1.2 V.

in the first cycle of scanning decreases and almost vanishes after several scanning cycles [31, 32].

2.5. Fabrication of the electrode materials

For the fabrication of electrodes, active materials (E-RGO or C-RGO) (prepared separately), conductive carbon black C65, and a 60% solution of PTFE (Polytetrafluoroethylene) were thoroughly mixed in 90:5:5 weight ratio. The mixture of materials was ground in an agate mortar to get a homogenous dough-like material. The black electrodes were rolled out whilst periodically adding 90% ethanol. They were then dried in an oven at 100 °C for 2-3 h to allow the alcohol to evaporate completely.

After that, the electrodes were cut in 10 mm discs, and their masses were approximately: E-RGO = +3.7 mg/-3.9 mg; C-RGO = +4.1 mg/-4.1 mg. A symmetric Swagelok-type 2E cell was assembled using a glass fiber separator (Whatman GF/A) prewetted with 1 M NaNO₃ aqueous electrolyte.

2.6. Electrochemical testing formulas

Electrochemical characteristics of E-RGO and C-RGO materials were measured using a potentiostat/galvanostat (SP150E BioLogic, France). Cyclic voltammetry (CV) and Galvanostatic charge-discharge (GCD) were performed for both of the electrode materials in 5 various potential windows (starting from 0 to 0.7; 0.9; 1.1; 1.3 and 1.5 V) at a 2 mV·s⁻¹ scan rate.

From the CV (1) and GCD (2) profiles the specific charge capacitance (F·g⁻¹) was calculated:

$$C_{CV} = \frac{2 \cdot A}{m_{total} \cdot v \cdot \Delta V} \quad (1)$$

$$C_{GCD} = \frac{4 \cdot I \cdot \Delta t}{\Delta V \cdot m} \quad (2)$$

where: A – integrated area of the CV lines; I – discharge current, A; ΔV – the potential range with the exception of IR drop (V); m – mass of the both active materials; v – scan rate, V·s⁻¹; Δt – the discharge time.

The Energy efficiency (η_E, %) (3) and Coulombic efficiency (η_i, %) (4) were calculated using following equations:

$$\eta_E = \frac{E_{int/D}}{E_{int/G}} \cdot 100\% \quad (3)$$

$$\eta_i = \frac{t_D}{t_G} \cdot 100\% \quad (4)$$

where: E_{int/D} – integral of discharging, V·s; E_{int/G} – integral of charging, V·s; t_D – time of discharging, s; t_G – time of charging, s.

3. Results and Discussion

3.1. Structural analysis of GO synthesis by XRD and SEM/TEM

As shown in Fig. 4, XRD pattern obtained in a range of 5° to 70°. This result confirms the successful synthesis of GO via the modified Hummers' method. A strong diffraction peak is observed at 2θ ≈ 9.8°,

which is assigned to the (001) reflection of GO [33]. According to Waheed Gul et al., the spectrum also displays a peak at 2θ = 12.83°, indicating that the GO peak corresponds to the 2θ range of 9–13° [34]. One more peak was observed at about 18° corresponding to an interlayer spacing of about 0.34 nm [35]. These two peak indicate the distance between graphene layers.

Also, XRD technique confirms that d-spacing is 0.89, which is characteristic of GO. At 25°, the peak starts to disappear because of the formation of oxygen containing groups, such as hydroxyl, epoxy, carbonyl, and carboxyl groups.

The production of GO was confirmed by a color change from black to brown during the oxidation process. The resulting material dispersed easily in water, forming a stable colloidal suspension [8]. This indicates successful exfoliation. Figure 5 provides a clear view of the morphology of the synthesized graphite oxide from both TEM and SEM analyses. In the TEM image (Fig. 5a), the material appears as thin, sheet-like structures with a transparent and wrinkled texture. The transparency suggests that the sheets are only a few layers thick, while the folds and overlaps indicate a flexible, exfoliated structure. These wrinkles are commonly observed in GO and are typically caused by the disruption of the carbon lattice during oxidation. The SEM image (Fig. 5b) shows a more compact and layered surface with a slightly rough and crumpled appearance. The sheets appear to be stacked or aggregated, which is likely due to restacking during sample preparation or drying. Overall, both images confirm the formation of thin, exfoliated GO sheets with a characteristic wrinkled morphology, along with some degree of restacking.

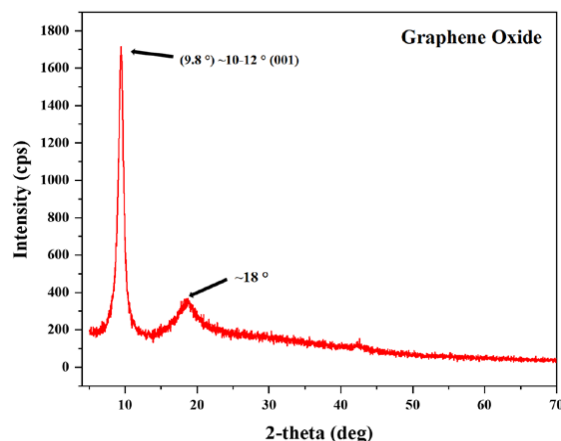


Fig. 4. XRD analysis of synthesized GO by using modified Hummer's method.

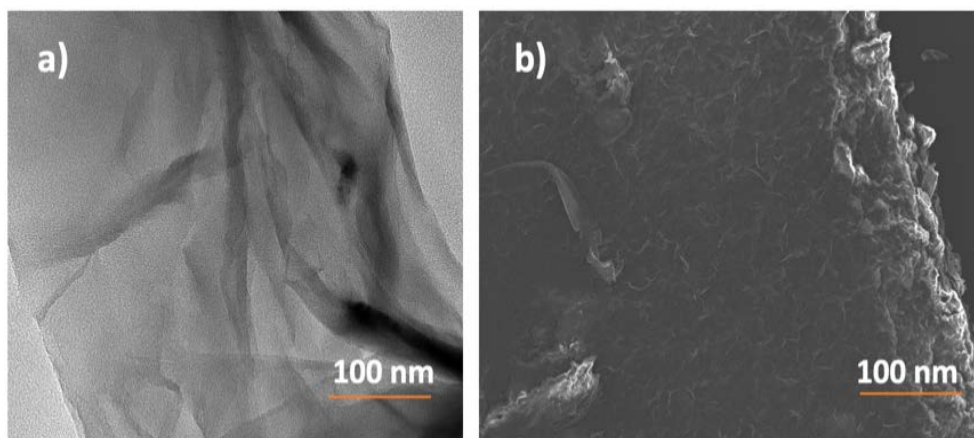


Fig. 5. TEM (a) and SEM (b) images of the graphene oxide after oxidation of graphite.

3.2. Structural characterization by Raman spectroscopy

The Raman spectra of the C-RGO and E-RGO presented in Fig. 6, demonstrating structural characteristics. Both reduced samples exhibit D and G bands. For E-RGO, the D and G bands are located at approximately 1344 and 1575 cm^{-1} , respectively, whereas C-RGO exhibits the D and G bands at approximately 1341 and 1581 cm^{-1} , respectively. The D band is associated with defects and disorder in the sp^2 carbon structure, while the G band corresponds to the in-plane vibration of sp^2 -bonded carbon atoms [19].

The ratio was used to evaluate the degree of structural disorder. The calculated values of I_D/I_G were 0.85 for E-RGO and 0.84 for C-RGO. The similar ratios indicate a comparable degree of structural disorder in both reduced materials and the difference between the two samples was minor.

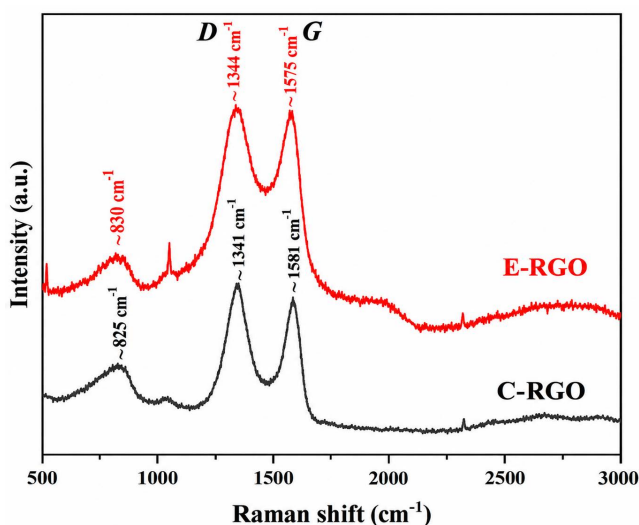


Fig. 6. Raman spectra showing the D and G bands of the reduced graphene oxide electrodes, C-RGO and E-RGO.

3.3. SEM analysis of the surface morphology

The lateral dimensions of the flakes were measured from SEM micrographs obtained using a field-emission scanning electron microscope (FE-SEM, Thermo Scientific Quattro S, Thermo Fisher Scientific, USA).

The micrograph for graphite (Fig. 7a) shows large, stacked plate like particles with well defined, flat basal faces. After oxidative exfoliation (Fig. 7b), GO appear thin and highly wrinkled, with a characteristic «paper like» texture. The sheets are loosely stacked, producing a porous network of overlapping edges. SEM observations revealed that the oxidized graphite retained lateral dimensions in the range of 1-5 μm . The surface morphology became highly distorted with visible waviness and sheet deformation, which can be attributed to the introduction of oxygenated surface species including hydroxyl, epoxy, and carboxyl moieties. Fig. 7c shows electrochemically reduced material in an aqueous electrolyte. The reduced materials exhibit smoother and flatter surfaces compared to graphene oxide, yet retain some wrinkling. Chemical reduction (Fig. 7d) using ascorbic acid leads to the formation of deformed flakes. The edges of materials reduced by chemical and electrochemical means appear to have more distinct boundaries. These structural changes suggest partial elimination of oxygenated species together with restoration of sp^2 -hybridized carbon domains.

All four samples exhibit lateral dimensions of approximately 1-5 μm , but the degree of exfoliation, surface smoothness, and stacking vary markedly as a function of the reduction pathway.

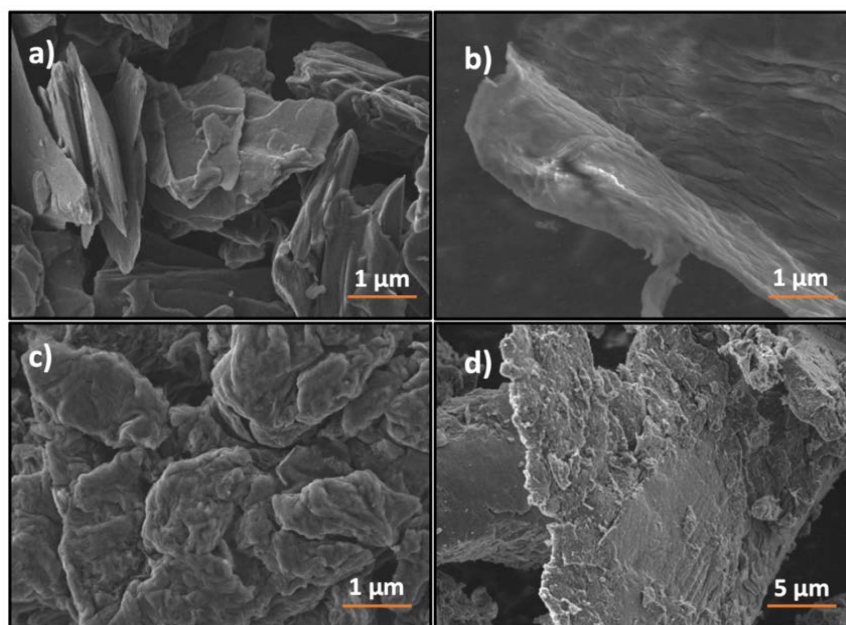


Fig. 7. SEM images at the same 1 μm scale of graphite (a), exfoliated graphene oxide (b), E-RGO (c), C-RGO (d); all materials have particle sizes ranging from 1 to 5 μm.

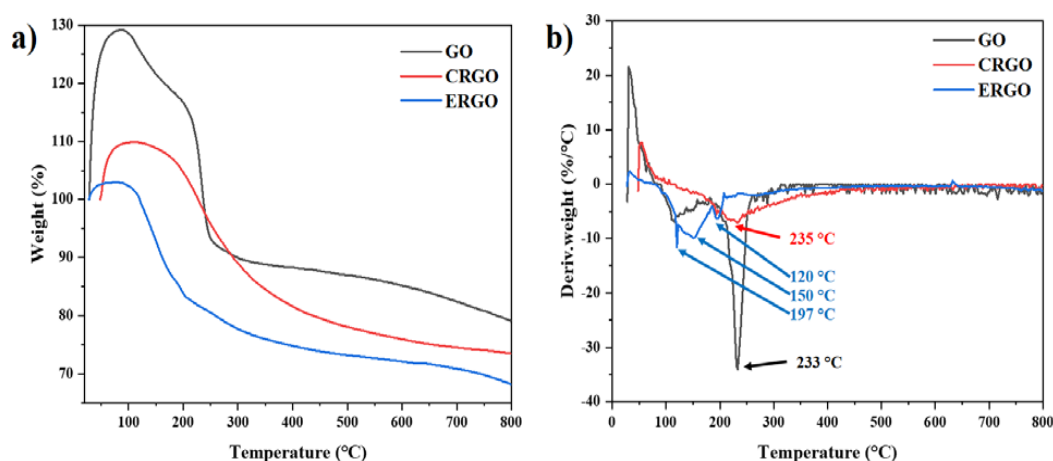


Fig. 8. TGA (a) and dTG (b) curve lines of GO, C-RGO, and E-RGO under N₂ atmosphere.

3.4. Thermogravimetric analysis (TGA)

The thermogravimetric analysis (TGA) and derivative thermogravimetric analysis (dTG) were conducted to compare the thermal stability and mass change rate of the samples as a function of temperature (28–48 °C to 800 °C) (Fig. 8a, b) in an inert N₂ atmosphere. TGA and dTG were performed using a NETZSCH STA 409 PC Luxx thermal analyzer at Satbayev University (Mining and Metallurgical Building, Almaty, Kazakhstan).

For all three samples, mass loss (%) was observed. GO initially showed an increase in mass percentage in the temperature range from 28 °C to 88 °C, reaching approximately 30% compared with the initial mass. Then, starting from 90 °C, gradual

mass loss began. At 208–215 °C, a sharp decrease occurred, and graphene oxide lost approximately 22% of its mass.

In contrast, CRGO and ERGO samples showed multistep weight-loss characteristics. In the case of CRGO, the mass increased by up to 10% at 110 °C, while no mass increase was observed for ERGO. CRGO started to lose mass at ~130 °C, while ERGO started to lose mass at ~90 °C, which can be attributed to the removal of physically adsorbed H₂O in the temperature range of 100–150 °C [37]. Usually, in the temperature range of 150–300 °C, the remaining oxygen-containing functional groups decompose, accompanied by the release of H₂O and CO₂ [38]. Further, a gradual mass loss was observed for all three samples upon heating up to 800 °C.

In Fig. 8b, the main DTG peak of GO was observed at approximately 233 °C, corresponding to the rapid decomposition of oxygen-containing functional groups. C-RGO exhibited a much less intense peak at approximately the same temperature, indicating a significant decrease in the amount of these functional groups after chemical reduction. In contrast, E-RGO showed no pronounced decomposition peak at 233–235 °C, displaying only several weak features at approximately 120, 152 and 197 °C. The substantially reduced DTG intensity of E-RGO indicates a lower content of thermally labile oxygen-containing groups and suggests a more effective removal of these functionalities during electrochemical reduction.

3.5. X-ray photoelectron spectroscopy analysis

The reduced samples and precursors were further analyzed by X-ray photoelectron spectroscopy (XPS) to evaluate the elemental composition of C and O. In Fig. 9, all four materials demonstrate mainly the presence of C and O atoms. However, a marginal amount of potassium detected in E-RGO (Fig. 10d), which was a result of the electrochemical reduction process.

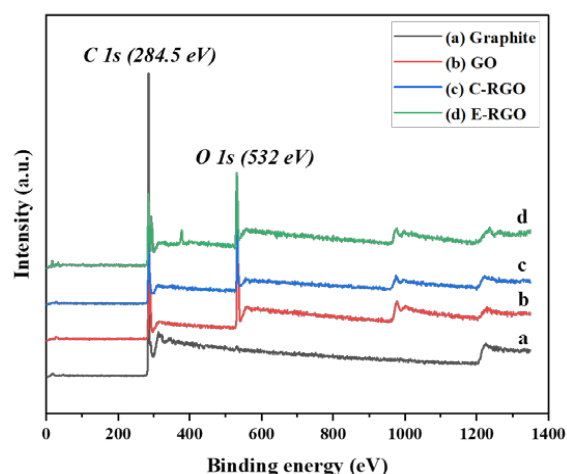


Fig. 9. XPS survey spectra of graphite (a), GO prepared via Hummers' method (b), C-RGO prepared using ascorbic acid (c) and E-RGO reduced in KOH electrolyte (d).

High-resolution C 1s spectra reveal that graphite is dominated by the C–C/C=C peak located at ~284.5 eV [39]. After the Hummers' treatment with strong oxidizing agents, the carbon atomic mass of GO decreased to 50.35 at.%, while oxygen content in O 1s region increased to 25 at.% with the presence of C–O ~286.4 eV; C=O ~287.5 eV; O–C=O ~288.5 eV, which

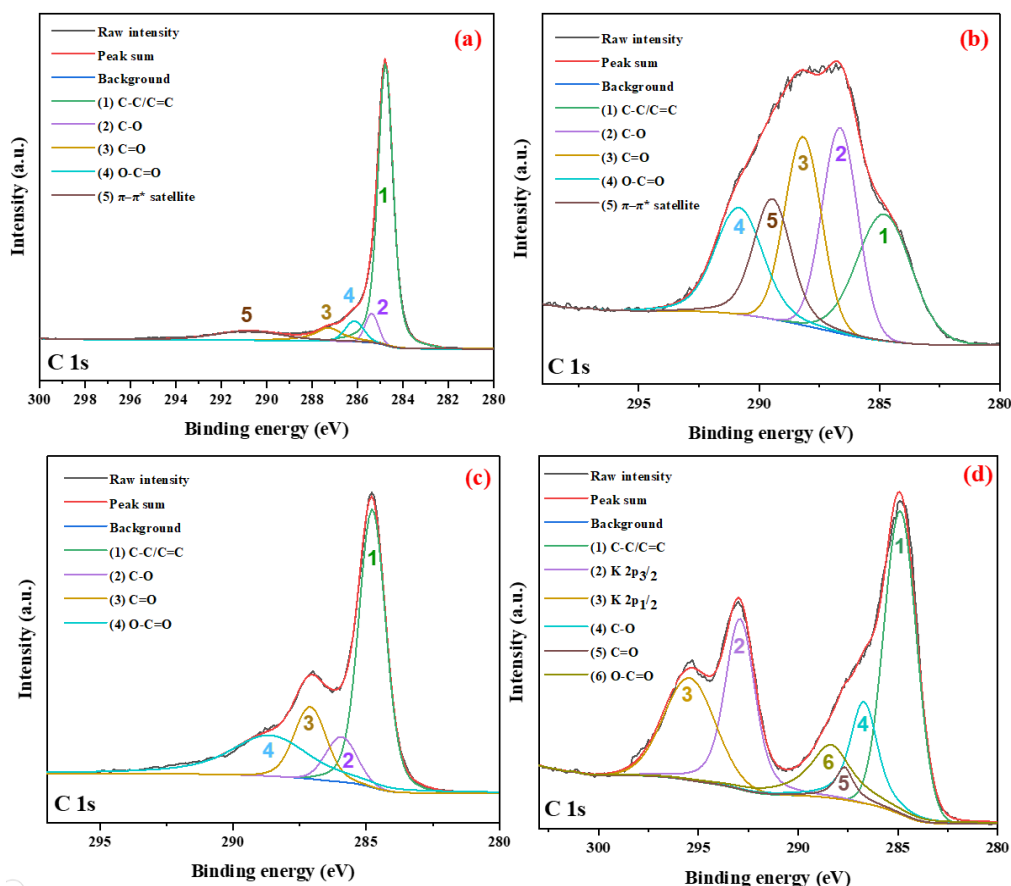


Fig. 10. Typical high-resolution XPS spectra of C 1s region of graphite (a), GO (b), C-RGO (c) and E-RGO (d).

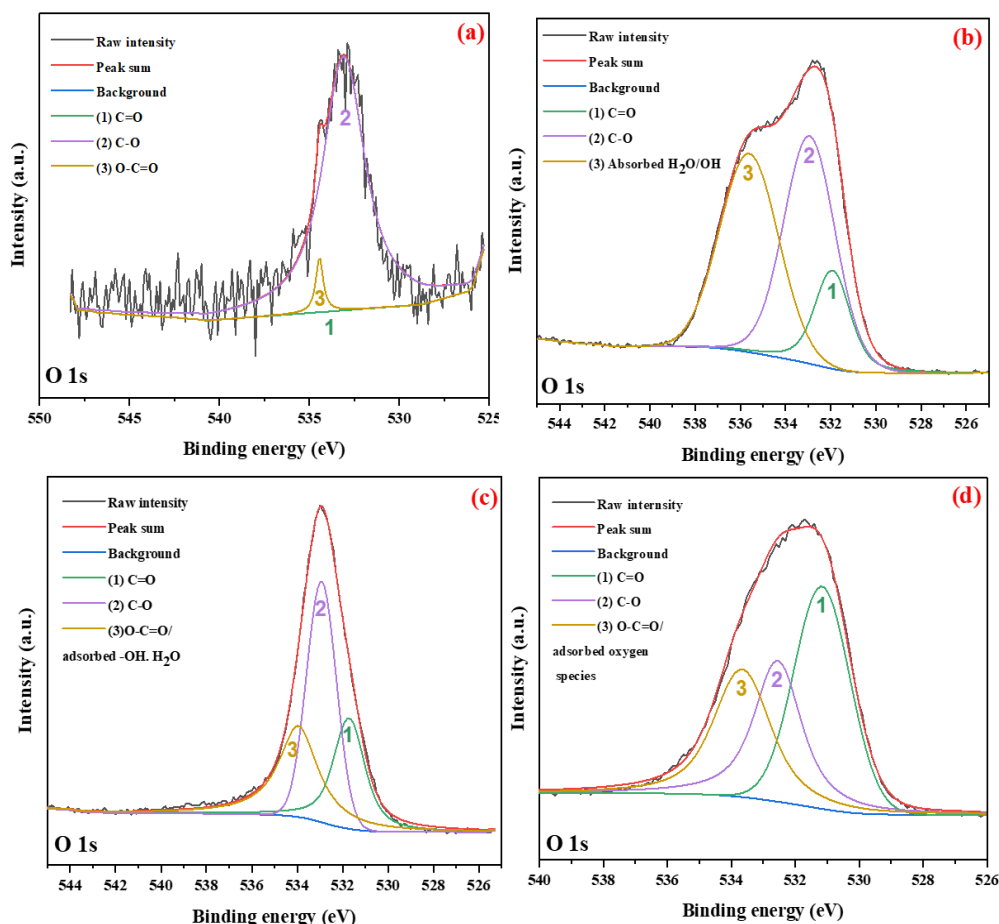


Fig. 11. Typical high-resolution XPS spectra of O 1s region of graphite (a), GO (b), C-RGO (c) and E-RGO (d).

proves about good oxidation process. As shown in Fig. 11b-11d, after the reduction process, the oxygen content in O 1s areas decreased in both reduced oxide samples, indicating the removal of the oxygen containing functional groups [40]. The oxygen content decreased from 25 at.% to 23.15 at.% and 17 at.% in C-RGO and E-RGO, respectively. The lower oxygen content in E-RGO than C-RGO suggests that electrochemical reduction method is more effective approach of removing oxygen-containing groups. The observation with the C 1s spectra, which show a higher C–C/C=C component and a lower contribution of oxygen-containing functional groups in E-RGO. In addition, the high-resolution C 1s spectrum of E-RGO showed the presence of potassium species K 2p_{3/2} (~292.9 eV) and K 2p_{1/2} (~295.6 eV), which are attributed to residual potassium originating from the KOH electrolyte used during the electrochemical reduction process.

3.6. Electrochemical characterizations of E-RGO and C-RGO

The electrochemical characteristics of the E-RGO and C-RGO electrodes were evaluated using a

two-electrode (2E) Swagelok cell configuration. An aqueous 1 M NaNO₃ solution was employed as the electrolyte for CV (Fig. 12a, b) and galvanostatic charge-discharge (GCD) (Fig. 13a, 13b) testing for both reduced materials.

Figure 12a shows the cyclic voltammetry curves of the electrochemically reduced E-RGO electrode. The corresponding cyclic voltammetry curves obtained at a scan rate of 2 mV·s⁻¹ over different potential windows; active mass of both E-RGO electrodes equal to 6.84 mg. The operating voltage window was initially from 0 to 0.7 V and subsequently extended to 0.9; 1.1; 1.3 and 1.5 V. As shown in the Fig. 12, when the voltage range is expanded from 0.7 to 1.5 V CV-profiles shift toward a more rectangular shape of the CV curves is observed, accompanied by an increase in current density, which indicates an improvement in electrochemical capacitive characteristics. The current density in CV was normalized to the mass of the active material (mA·g⁻¹). For potential ranges from 0 to 0.7 V (black line), the current density area is 32.4 mA·g⁻¹; from 0 to 0.9 V (red line), the current density area is 35.6 mA·g⁻¹; from 0 to 1.1 V (blue line), the current density area is 39.8 mA·g⁻¹.

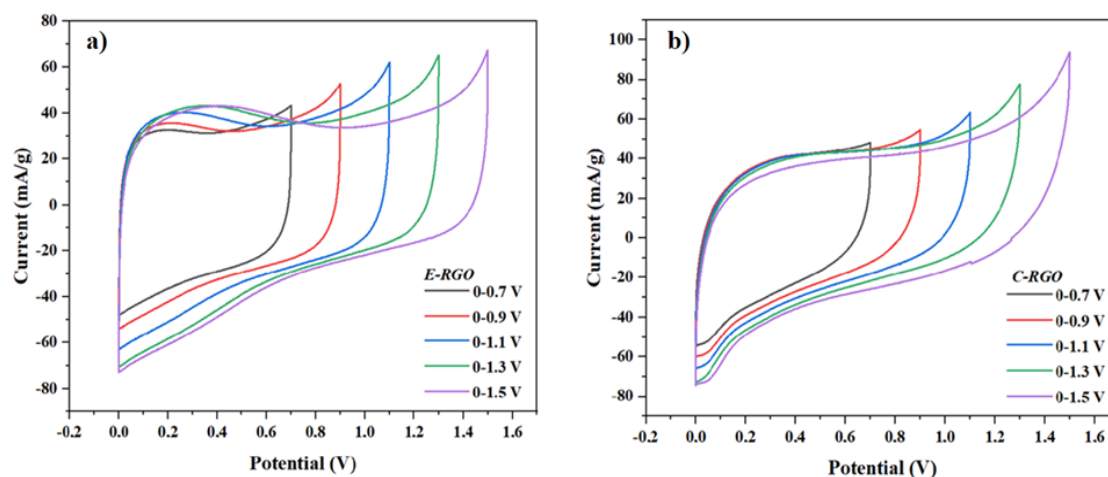


Fig. 12. CV curves of electrodes prepared from E-RGO (a) and C-RGO (b) were recorded in a Swagelok-type 2E cell using 1 M NaNO_3 as the electrolyte; scan rate of $2 \text{ mV}\cdot\text{s}^{-1}$ in different potential ranges.

For wider ranges up to 1.3 and 1.5 V (green and purple lines), the current density area is 42.5 and $43.1 \text{ mA}\cdot\text{g}^{-1}$, respectively.

Figure 12b shows the cyclic voltammetry curves of the chemically reduced C-RGO electrode. Graphene oxide powders obtained via the Hummers' method from pure graphite were chemically reduced using 1 M ascorbic acid. Just as with E-RGO, C-RGO electrodes were scanned at a rate of $2 \text{ mV}\cdot\text{s}^{-1}$ across various potential ranges. Calculations show that the current density per unit area for the potential range up to 0.7 V is $27.9 \text{ mA}\cdot\text{g}^{-1}$; up to 0.9 V, it is $34.8 \text{ mA}\cdot\text{g}^{-1}$; up to 1.1 V, it is $37.2 \text{ mA}\cdot\text{g}^{-1}$; for 1.3 and 1.5 V, they are 37.9 and $38.5 \text{ mA}\cdot\text{g}^{-1}$, respectively.

The results show nearly identical current-area values, with differing values for the electrochemically reduced material compared to the chemically reduced material.

GCD characteristics of the E-RGO and C-RGO electrodes (Fig. 13a, 13b) recorded in 1 M Sodium nitrate at progressively expanded potential windows (0–0.7 V to 0–1.5 V) provide insight into its charge storage mechanism and operational stability. As the potential window is extended beyond 1.1 V, a noticeable increase in discharge time is observed, reflecting enhanced charge storage and higher apparent capacitance [41]. The reduced symmetry at higher voltages further suggests a decline in coulombic efficiency, likely due to irreversible electrochemical reactions [42]. Fig. 13a shows a GCD curve for electrodes made from E-RGO material.

To calculate the specific charge capacitance, the following data were obtained, along with the calculation formula used to derive the final capacitance value for each potential range.

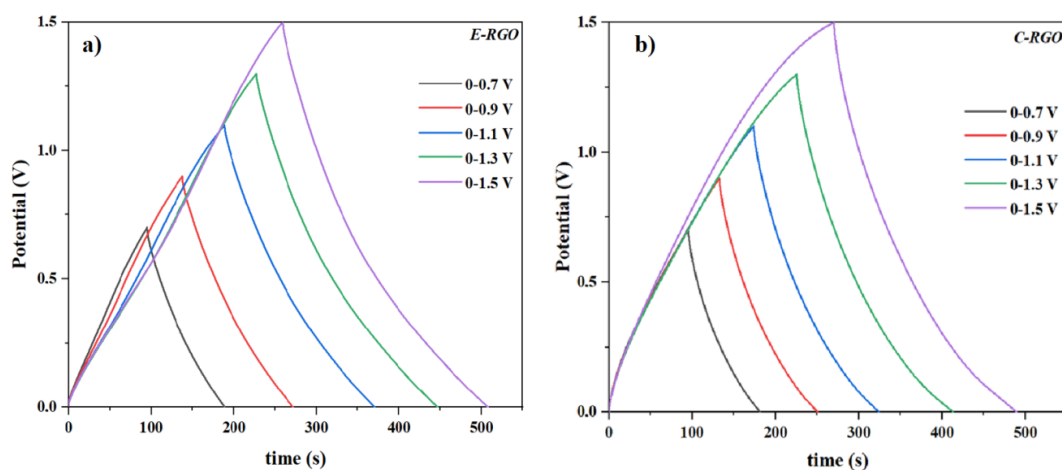


Fig. 13. Electrochemical behavior of E-RGO (a) and C-RGO (b) in a Swagelok Type 2E cell using GCD in a 1 M NaNO_3 electrolyte at a scan rate of $2 \text{ mV}\cdot\text{s}^{-1}$.

According to the study by Jiang Yang et al. [18], electrochemical characterization of E-RGO by cyclic voltammetry at a scan rate of $5\text{ mV}\cdot\text{s}^{-1}$ yielded a specific capacitance of $223.6\text{ F}\cdot\text{g}^{-1}$. This value is higher than that obtained in the present study, where the specific capacitance calculated from the GCD curves increased from $54\text{ F}\cdot\text{g}^{-1}$ in the voltage range of 0-0.7 V to $67\text{ F}\cdot\text{g}^{-1}$ in the extended voltage range of 0-1.5 V (Table 1). However, a direct comparison of these results is not appropriate because the electrochemical testing conditions differ significantly. In the present study, cyclic voltammetry was performed at a scan rate of $2\text{ mV}\cdot\text{s}^{-1}$, whereas Jiang Yang et al. employed a scan rate of $5\text{ mV}\cdot\text{s}^{-1}$. Furthermore, different electrolytes were used: 1 M NaNO_3 in the present work and 1 M H_2SO_4 in the referenced study, which may also have a significant influence on the specific capacitance. In addition, the energy efficiency and Coulombic efficiency of the assembled device were 72 and 102%, respectively (Table 2).

In the case of C-RGO, according to the work by Kai Zhang et al. [45], the highest specific capacitance is $194\text{ F}\cdot\text{g}^{-1}$ at a current density of $1\text{ A}\cdot\text{g}^{-1}$ in a 2 M H_2SO_4 electrolyte. In this experimental study, for chemically reduced graphene oxide, the specific capacitance (Table 3) calculated from the GCD curves

is $50\text{ F}\cdot\text{g}^{-1}$ in the potential range of 0-0.7 V and gradually increases to $59\text{ F}\cdot\text{g}^{-1}$ when the operating range is expanded to 0-1.5 V. In this study, 1 M NaNO_3 was used as the electrolyte, and electrochemical measurements were performed at a scan rate of $2\text{ mV}\cdot\text{s}^{-1}$. The energy efficiency (Table 4) was 55%, and the Coulombic efficiency was 102%. Based on the shapes of the CV curves and the specific capacitance values, it can be concluded that the electrochemical method of graphene oxide reduction provides higher electrochemical activity compared to the chemical reduction method. Furthermore, electrochemically reduced graphene oxide is characterized by a higher specific capacitance. One of the main advantages of electrochemical reduction is the presence of distinct reduction peaks on the CV curves, which indicates that the reduction process is taking place and allows it to be monitored directly during the electrochemical experiment.

Figure 14 presents the Nyquist plots of E-RGO and C-RGO together with the corresponding equivalent electrical circuit (EEC) model, as $R_s - R_c \parallel C_{dl}$. Here, R_s – solution resistance, R_{ct} – charge transfer resistance, C_{dl} – double layer capacitance.

The EIS measurements were performed in 1 M NaNO_3 electrolyte. The Nyquist plots of E-RGO and

Table 1. Obtained charge and discharge time for E-RGO electrodes from GCD curves to calculate specific charge capacitance, F g^{-1} from the Formula 1

Applied potential range, V	Time of charging, s	Time of discharging, s	IR drop, s	V max, V	Δt	Specific charge capacitance, $\text{F}\cdot\text{g}^{-1}$
0-0.7 V	10 853.6	11 042.3	10 948.7	0.693	93.6	54
0-0.9 V	22 501.8	22 773.9	22 639.6	0.893	134.3	60
0-1.1 V	37 011.7	37 382.2	37 200.3	1.094	181.9	67
0-1.3 V	54 331.1	54 777.9	54 558.4	1.296	219.5	68
0-1.5 V	74 288.8	74 796.9	74 548.0	1.495	248.9	67

Note: I (Current) – $0.684\text{ mA} = 0.000684\text{ A}$ (constant for all potential ranges); m (Active mass for both electrodes) – $6.84\text{ mg} = 0.00684\text{ g}$ (constant for all potential ranges); Δt – discharge time; ΔV – voltage window.

Table 2. Energy efficiency (η_E , %) and Coulombic efficiency (η_t , %) for E-RGO

Applied potential range, V	Time of charging, s	Time of discharging, s	Integral of charging, V.s ($E_{int/D}$)	Integral of discharging, V.s ($E_{int/D}$)	Energy efficiency, % (η_E)	Coulombic efficiency, % (η_t)
0-0.7 V	10 853.6	11 042.3	36.47	25.95	72	102
0-0.9 V	22 501.8	22 773.9	67.04	47.60	71	101
0-1.1 V	37 011.7	37 382.2	109.67	76.53	70	101
0-1.3 V	54 331.1	54 777.9	151.13	105.52	70	101
0-1.5 V	74 288.8	74 796.9	197.58	134.37	68	101

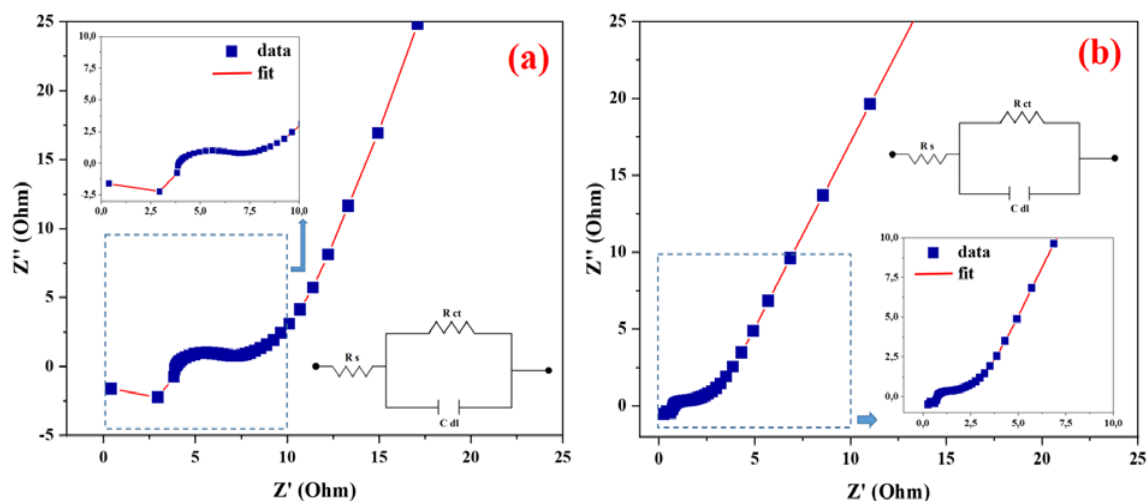
Table 3. Obtained charge and discharge time for C-RGO electrodes from GCD curves to calculate specific charge capacitance, $F \cdot g^{-1}$

Applied potential range, V	Time of charging, s	Time of discharging, s	IR drop, s	V max, V	Δt	Specific charge capacitance, $F \cdot g^{-1}$
0-0.7 V	10 819.2	11 001.1	10 914.5	0.699	86.6	50
0-0.9 V	22 356.0	22 606.7	22 488.7	0.900	118.0	53
0-1.1 V	36 603.6	36 928.1	36 777.3	1.099	150.8	55
0-1.3 V	53 647.1	54 060.5	53 872.5	1.299	188.0	58
0-1.5 V	73 455.0	73 944.6	73 725.1	1.499	219.5	59

Note: I (Current) – 0.738 mA = 0.000738 A (constant for all potential ranges); m (Active mass for both electrodes) – 7.38 mg = 0.00738 g (constant for all potential ranges); Δt – discharge time; ΔV – voltage window.

Table 4. Energy efficiency (η_E , %) and Coulombic efficiency (η_t , %) for C-RGO

Applied potential range, V	Time of charging, s	Time of discharging, s	Integral of charging, V.s ($E_{int/D}$)	Integral of discharging, V.s ($E_{int/D}$)	Energy efficiency, % (η_E)	Coulombic efficiency, % (η_t)
0-0.7 V	10 819.2	11 001.1	39.38	21.80	55	102
0-0.9 V	22 356.0	22 606.7	69.26	37.68	54	101
0-1.1 V	36 603.6	36 928.1	110.43	57.89	52	101
0-1.3 V	53 647.1	54 060.5	173.27	82.95	48	101
0-1.5 V	73 455.0	73 944.6	250.20	109.53	44	101

**Fig. 14.** Nyquist plots (EIS) of E-RGO (a) and C-RGO (b) electrodes in the voltage range: 0-0.7 V, together with the equivalent electrical circuit (EEC) model.

C-RGO exhibit almost the same real-axis intercept (Fig. 14a, 14b), indicating similar equivalent series resistance (ESR). The intercept of the Nyquist plot with the real impedance axis (Z') at high frequency is associated with the R_s , which includes the resistance of the electrolyte, electrode material, current collector, and electrical contacts.

In the present EDLC system, C_{dl} describes the electrostatic accumulation of ions at the electrode/electrolyte interface, while R_{ct} reflects the resistance associated with interfacial charge transport and ion accessibility. At lower frequencies, the increase in the imaginary impedance and the formation of a steeply inclined line indicate predominantly capac-

itive behavior and ion transport within the porous electrode structure. The experimental impedance data were fitted using the proposed EEC model to evaluate the resistive and capacitive contributions of the E-RGO and C-RGO electrodes and concluded that both reduced material display low internal resistance and favorable ion-transport characteristics, supporting their suitability for EDLC applications.

4. Conclusion

The electrochemical treatment of graphene oxide on stainless-steel foil provides a rapid and scalable route for producing conductive reduced graphene oxide. Under a mild cathodic potential (vs. Ag/AgCl) in a three-electrode system, oxygen-containing groups were efficiently removed, while XRD analysis confirmed effective deoxygenation together with partial reconstruction of the graphitic lattice. The resulting E-RGO exhibits a marked increase in the C/O atomic ratio and an electrical conductivity of ≈ 1 S/m, substantially higher than that of chemically reduced analogues. Electrochemical reduction in 1 M KOH introduced clean reduction peaks (-1.1 V; -0.8 V), which indicate ER as controllable reduction technique. Electrochemical performance testing shows good capacitance and cycle stability for both E-RGO and C-RGO. At the result, E-RGO (67 F·g $^{-1}$) materials show 13.7% better electrochemical characteristics than C-RGO (59 F·g $^{-1}$). Overall, the study demonstrates that electrochemical reduction is an environmentally good, controllable alternative to hazardous chemical reducers, enabling the production of E-RGO with reproducible structural and electrochemical properties. Future work should explore electrolyte engineering and in situ monitoring to further optimize reduction depth and tailor surface functionalities for specific device architectures.

Author Contributions

Zhazira Supiyeva – Conceptualization, Supervision, Resources, Funding Acquisition, Project Administration, Writing – Review & Editing. **Balaussa Balgabayeveva** – Methodology, Investigation, Validation, Writing – Original Draft, Writing – Review & Editing. **Nurbatyr Mukhametgazy** – Methodology, Formal Analysis. **Fail Sultanov** – Investigation, Writing – Review & Editing. **Aigul Tugelbayeva** – Data Curation, Visualization. **Xuexue Pan** – Supervision, Validation, Writing – Review & Editing

Conflict of Interest

The authors declare that they have no conflict of interest.

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Синтез и характеристика электрохимически восстановленного оксида графена для применения в суперконденсаторах

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АННОТАЦИЯ

Оксид графена (GO) получен из графита модифицированным методом Хаммерса с использованием сильных окислителей, после чего он был восстановлен путем удаления кислородсодержащих функциональных групп. sp^2 -углеродные домены в материале GO не обеспечивают перенос электронов из-за плотной геометрии и более высокой электроотрицательности sp^2 -орбиталей по сравнению с sp^3 -углеродными центрами. Успешный синтез GO из графитового порошка подтвержден методом рентгеновской дифракции (XRD), показавшим дифракционный пик при $2\theta \sim 9,8^\circ$. Далее процедуры восстановления GO проводили электрохимическим (трех-электродная ячейка, 1 М КОН, диапазон потенциала от 0 до -1,2 В) и химическим (1 М аскорбиновая кислота, 60 °С, перемешивание) методами. Морфологический анализ (сканирующая электронная микроскопия (SEM) и просвечивающая электронная микроскопия (ТЕМ)), структурный анализ (рентгеновская фотоэлектронная спектроскопия (XPS) и рамановская спектроскопия) и термический анализ (термогравиметрический анализ (TGA)) подтвердили удаление кислородсодержащих групп, указывая на превращение GO в восстановленный GO (rGO). Электрохимические характеристики (CV, GCD, EIS) исследованы в симметричной двухэлектродной ячейке типа Swagelok с использованием 1 М NaNO_3 . На основании полученных результатов наиболее перспективным материалом оказался электрохимически восстановленный оксид графена (E-RGO), для которого удельная емкость увеличилась с $\sim 54 \text{ Ф} \cdot \text{г}^{-1}$ при 0,7 В до $\sim 67 \text{ Ф} \cdot \text{г}^{-1}$ при 1,5 В при удельной плотности тока $200 \text{ мА} \cdot \text{г}^{-1}$ в электролите 1 М NaNO_3 . Удельная емкость электродов E-RGO на 13,7% выше, чем у химически восстановленного оксида графена (C-RGO), и эти результаты демонстрируют эффективность электрохимически восстановленного оксида графена, дополнительно подтверждая его потенциальное применение в суперконденсаторах.

Ключевые слова: оксид графена, восстановленный оксид графена, электрохимическое восстановление, циклическая вольтамперометрия, суперконденсаторы, хранение энергии

Суперконденсаторларда қолдануға арналған электрохимиялық тотықсыздандырылған графен оксидін синтездеу және сипаттау

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АҢДАТПА

Графен оксиді (GO) графиттен күшті тотықтырғыштарды қолдану арқылы модификацияланған Хаммерс әдісімен алынды. Кейін оның құрамындағы оттеқ-құрамды функционалдық топтарды жою арқылы тотықсыздандырылды. GO материалындағы sp^2 -көміртекті домендер тығыз геометрияға және sp^3 -көміртеқ орталықтарымен салыстырғанда sp^2 -орбитальдардың электртерістілігінің жоғары болуына байланысты электрондардың тасымалдануына мүмкіндік бермейді. Графит ұнтағынан GO-ның сәтті синтезделуі рентгендік дифракция (XRD) әдісімен расталды, мұнда $2\theta = \sim 9,8^\circ$ мәнінде дифракциялық шың байқалды. Одан әрі GO-ны тотықсыздандыру процестері электрохимиялық (үш электродты ұяшық, 1 М КОН, потенциал диапазоны 0-ден -1,2 В-қа дейін) және химиялық (1 М аскорбин қышқылы, 60 °С, араластыру) әдістер арқылы жүргізілді. Морфологиялық (сканерлеуші электрондық микроскопия (SEM) және трансмиссиялық электрондық микроскопия (TEM)), құрылымдық (рентгендік фотоэлектрондық спектроскопия (XPS) және раман спектроскопиясы) және термиялық (термогравиметриялық талдау (TGA)) талдаулар оттеққұрамды топтардың жойылғанын растады, бұл GO-ның тотықсыздандырылған GO-ға (rGO) айналғанын көрсетеді. Электрохимиялық сипаттамалар (CV, GCD, EIS) 1 М NaNO₃ қолданылған симметриялық екі электродты Swagelok типті ұяшықта зерттелді. Алынған нәтижелерге сүйене отырып, ең перспективалы материал электрохимиялық жолмен тотықсыздандырылған графен оксиді (E-RGO) болды, оның меншікті сыйымдылығы 1 М NaNO₃ электролитінде 200 мА·г⁻¹ меншікті ток тығыздығында 0,7 В кезінде ~ 54 Ф·г⁻¹-ден 1,5 В кезінде ~ 67 Ф·г⁻¹-ге дейін артты. E-RGO электродтарының меншікті сыйымдылығы химиялық жолмен тотықсыздандырылған графен оксидімен (C-RGO) салыстырғанда 13,7% жоғары, және бұл нәтижелер электрохимиялық жолмен тотықсыздандырылған графен оксидінің тиімділігін көрсетіп, оның суперконденсаторларда қолданылу әлеуетін одан әрі дәлелдейді.

Түйін сөздер: графен оксиді, тотықсыздандырылған графен оксиді, электрохимиялық тотықсыздандыру, цикликалық вольтамперометрия, суперконденсаторлар, энергия сақтау