p-ISSN 2089 – 0133 e-ISSN 2477-6416
URL: https://jurnal.uns.ac.id/ijap/article/view/88878

DOI: https://doi.org/10.13057/ijap.v15i1.88878

SYNTHESIS AND CHARACTERIZATION OF REDUCED GRAPHENE OXIDE (rGO) AS A SOLID-STATE ELECTROLYTE IN BATTERIES

Lela Mukmilah Yuningsih*, Devi Indah Anwar, Eva Fauziah
Chemistry Study Program, Faculty of Science and Technology, Universitas Muhammadiyah Sukabumi,
Sukabumi, Indonesia
*lelathea@ummi.ac.id

Received 24-06-2024, Revised 14-09-2024, Accepted 27-02-2025,
Available Online 01-04-2025, Published Regularly April 2025

ABSTRACT

Reduced Graphene Oxide (rGO) is a derivative of graphene that has undergone the removal or reduction of its oxygen groups. This study performed the synthesis and characterization of rGO as a solid-state electrolyte in batteries in order to assess the conductivity of rGO and its ability to store electrical currents and voltages. The rGO was synthesized by the Hummers method and the reduction of graphene oxide with a zinc reductor. Graphite oxide was first synthesized by oxidizing graphite with H_2SO_4 dan $KMnO_4$. This graphite oxide was ultrasonically exfoliated and treated with a Zn reductor to produce rGO in powdered form. The rGO was characterized with Fourier Transform Infrared Spectroscopy (FTIR), X-Ray Diffraction (XRD), Scanning Electron Microscope (SEM), an LCR-meter, and a Digital Multimeter. This identified a diffraction pattern in rGO with a 2θ of 25.85° and a d-spacing of 3.444 Å with 48.76 % crystallinity. FTIR observation on rGO exhibited O-H vibrations at a wavelength of 3431.6 cm^{-1}, C=O at 1722.43 cm^{-1}, C=C (1573.91 cm^{-1}), and C-O (1080.14 cm^{-1}). The rGO was observed to have a triple-layered morphology at 10000x magnification. Conductivity for the rGO was identified as 0.000357005 S/cm, while the generated voltage was 1.21 V with a current of 3.55 mA.

Keywords: Conductivity; Graphite; Hummers; Reduced Graphene Oxide; Solid-state electrolyte.

Cite this as: Yuningsih, Y. M., Anwar, D. I., & Fauziah, E. 2025. Synthesis and Characterization of Reduced Graphene Oxide (rGO) as a Solid-State Electrolyte in Batteries. *IJAP: Indonesian Journal of Applied Physics, 15*(1), 124-132. doi: https://doi.org/10.13057/ijap.v15i1.88878

INTRODUCTION

Electrolytes are generally made out of good electrical conductors, including carbon. Studied the ultrasonic synthesis of PANI-Celluloseas a battery electrolyte, resulting in a conductivity of 0.00312 S/cm, voltage of 1.31 Volts and a current 71.4 mA [1]. PANI-Celluloseas is polyaniline (PANI) bound to cellulose. Polyaniline is a conductive polymer often used in various electronic applications and composite materials due to its ability to conduct electricity. Cellulose is a polysaccharide found in the cell walls of plants and is frequently used in industry as a base material for various products. Synthesized a polyaniline-carbon composite through the polymerization of aniline in varied temperatures; this PANI-carbon composite exhibited a conductivity of 0.718 S/cm, voltage of 1.09 Volts and a current 19.74 mA at 800°C, while at 1000°C the conductivity was 1.1003 S/cm with a voltage of 1.15 Volt and a current 19.98 mA [2]. Used sulfuric acid hydrolysis to achieve a conductivity of 3.6013 x 10^{-2} S/cm and a voltage of 1.34 Volts and a current 83.4 mA [3]. These studies so far have only been able to produce voltages significantly below the 1.5V standard for conventional batteries [4] in order to overcome this limitation, it was decided to experiment with a highly conductive material, namely Reduced Graphene Oxide (rGO).

rGO is a nanomaterial derivative of graphene that has been observed to exhibit high conductivity. In basic terms, rGO is an oxide of graphene where its oxide functional groups have been reduced or removed [5-6]. has previously synthesized, characterized, and measured the conductivity of rGO, resulting in a conductivity of 2.906 S/cm. There has been some research into the applied uses of rGO, including as a supercapacitor [7], a biomedical material [8], lithium-sulfur cathode [9], and an antibacterial [10]. The synthesis of rGO begins with the chemical synthesis of graphene oxide (GO) by oxidizing graphite powder with a strong acid, which is known as the Hummers method. Once the graphite has been oxidized by H_2SO_4 and $KMnO_4$, the resulting graphite oxide is ultrasonically exfoliated to produce graphene oxide (GO), which is then reduced by a reductor substance (Zn) to obtain powdered rGO [11].

This study sought to synthesize and characterize rGO as a solid-state electrolyte for batteries through the Hummers method and the subsequent reduction of graphene oxide with a zinc reductor. The rGO product was characterized with FTIR, SEM, XRD, an LCR-meter, and a digital multimeter.

METHOD

Sample

The sample used for this study was extracted from the graphite core of a 2B pencil faber castell.

Materials

The materials needed were graphite, distilled water smart lab A-1078, 95-97% H_2SO_4 merck 1.00731.2500, $NaNO_3$ merck 1.006537.0500, $KMnO_4$ merck 1.05082.250, 30% H_2O_2 merck 1.07209.1000, 35% and 5% HCl merck 1.00317.2500, zinc powder merck 1.08789.0500, and Whatman filter paper merck 1001-090.

Equipment

The tools and equipment utilized in this study were an analytic scale, porcelain bowls, funnels, measuring flasks, volumetric pipettes, measuring beakers, spatulas, hot plates, ovens, pH meters, Erlenmeyer flasks, ice bath, centrifuge, ultrasonic cleaner, glass beakers, batteries, as well as LCR meter merck Hioki IM3533, digital micrometer merck DT-9205A, X-ray diffraction (XRD) merck Rigaku Miniplex, scanning electron microscopy (SEM) merck JEOL JSM-6510LA, and Fourier transform infrared sprectroscopy (FTIR) instruments merck Shimadzu type prestige 21.

Synthesis of Graphite Oxide

Graphite oxide was synthesized through the Hummers method. 1.2 grams of graphite and 2 grams of $NaNO_3$ were dissolved in 50 mL of 97% H_2SO_4 and stirred for 2 hours at a temperature of 0-6°C. 6 grams $KMnO_4$ was slowly added into the solution while maintaining a temperature below 14°C, and then the solution was further stirred for 2 hours at a controlled temperature of 30°C. 100 mL of distilled water was added slowly, keeping the temperature below 96°C. Then another 200 mL of distilled water was added, followed by 8 mL of 30% H_2O_2. The solution was centrifuged at 5000 rpm for 15 minutes and then rinsed with 5% HCL and distilled water until the pH was close to neutral, then filtered and dried at 110°C for 12 hours to produce sheets of graphite oxide [11-12].

Reduction of Graphene Oxide

0.4 grams of GO was dissolved in 40 mL of distilled water, then subjected to ultrasonic treatment for 150 minutes to exfoliate graphite oxide into graphene oxide. 0.8 grams of Zn and 10 mL of 35% HCl was then added into the solution and stirred for 1 hour. 10 mL of HCl 35% was subsequently added and stirred again for 30 minutes before being rinsed with 5% HCl and distilled water until the solution's pH was neutral. The rinsed precipitate was put into a small stainless-steel container and heated in an oven at 200 °C (18 hours) to produce rGO powder [11].

Characterization of Graphite, Graphite Oxide, and rGO

The characterization of the sample consisted of the characterization of functional groups through FTIR, diffraction patterns and crystallinity level with XRD, morphological analysis with SEM, and electrical conductivity with an LCR meter [3].

Scheme to determine conductivity

Sample preparation

Prepare rGO samples in the form of powders.

Dimmension Measurement

Measure the physical dimensions of the sample (length, width, and thickness) accurately using measuring instruments such as a micrometer.

Selection of Measurement Method

Place four electrodes on the sample, two electrodes are tested for current, and the other two electrodes are measured for voltage. Resistance is obtained from the ratio of voltage to current.

Place four electrodes on the sample, two electrodes are tested for current, and the other two electrodes are measured for voltage. Resistance is obtained from the ratio of voltage to current.

Conductivity Calculation

Conductivity (σ): Calculate conductivity using the formula:

$$\text{Conductivity } (\sigma) = \frac{G \text{ X } L}{A}$$

Where:

σ = Conductivity (S/cm)

G = Conductance (S)

L = Sample Thickness (cm)

A = Electrode plat area (cm^2)

Voltage and Current Measurement for rGO

This was performed by removing the electrolyte from a battery, stuffing the cavity full of rGO tamped down to maximum density, then resealing the battery and measuring the resulting voltage and current with a digital multimeter [3].

RESULTS AND DISCUSSION

Synthesis of Graphite Oxide

The synthesis of graphite oxide was performed with the Hummers method [11-12]. This process began with the oxidation of graphite by means of an acidic oxidizer. At first, graphite and $NaNO_3$ was dissolved in H_2SO_4 and stirred for 2 hours while the temperature was kept low at 0 - 6°C in order to prevent damage to the graphite oxide and avoid the formation of dangerous gases in the form of NO_2 through the following reaction: [13].

$$NaNO_{3\,(S)} + H_2SO_{4\,(l)} \longrightarrow NaHSO_4 + HNO_3$$

$$4HNO_3 \longrightarrow 4NO_2 + O_2 + 3H_2O$$

The solution turned black during this oxidation process. Afterwards, $KMnO_4$ was added slowly into it while maintaining a temperature under 14°C since $KMnO_4$ could become explosive at higher temperatures through the generation of Mn_2O_7. The addition of $KMnO_4$ changed the solution's color into a greenish hue along with an exothermic reaction. The reaction that took place in this oxidation phase can be described as follows: [13].

$$2KMnO_4 + H_2SO_4 \longrightarrow K_2SO_4 + Mn_2O_7 + H_2O$$

The solution was then stirred for 2 hours while maintaining a temperature of 30°C, turning a brownish color in the process. This color change indicated that the reaction had taken effect according to the following scheme: [13].

$$3Mn_2O_7 \longrightarrow 6MnO_2 + 3O_2 + O_3$$

To reduce the solution's acidity and salt content, H_2O was added slowly while maintaining a temperature below 98°C, turning the solution dark brown. Then 30% H_2O_2 30% was added to stop the oxidation, which changed the solution's color once more to yellow. The reaction involved in this phase is described as follows: [13].

$$2MnO_4^- + 5H_2O_2 + 6H^+ \longrightarrow 2Mn^{2+} + 5O_2 + 8H_2O$$

The solution was centrifuged at 5000 rpm for 15 minutes to separate its components, and then the precipitate was extracted and rinsed with 5% HCl to remove contaminants such as metals, salts, and other unwanted substances prior to being neutralized with distilled water. Once the sample was neutral, it was dried in an oven at 110°C for 12 hours to produce sheets of graphite oxide. The rinsing reaction can be described as follows: [13].

$$2MnO_4^- + 16HCl \longrightarrow 6\,Cl^- + 2Mn^{2+} + 5Cl_2 + 8H_2O$$

Reduction of Graphene Oxide

The rGO process was carried out with a zinc reductor. 0.4 grams of graphite oxide was dissolved in 40 mL of distilled water and stirred for 1 hour to homogenize the solution [11]. Ultrasonic treatment was performed for 150 minutes to disperse or exfoliate the graphite oxide solution into graphene oxide with lower particle sizes. This exfoliation took place mechanically through interactions with ultrasonic waves and cavitation processes, which turned graphite oxide into graphene oxide [14]. The reactions could be described as follows: [6].

$$Zn + 2H_2O \longrightarrow Zn\,(OH)_2 + 2H^+ + 2e^-$$
$$Zn\,(OH)_2 \longrightarrow ZnO + H_2O$$
$$GO + 2H^+ + 2e \longrightarrow rGO$$

35% HCl 35% and 0.8 grams of Zn was added slowly to the resulting solution, producing gas bubbles that indicated the ongoing oxidation process was eliminating imperfectly reduced oxygen groups.

X-Ray Diffraction (XRD) Analysis

XRD analysis was performed to find out the sample's diffraction patterns and level of crystallinity. Observations were made with X-ray diffraction at angles of 2θ= 5° - 90°. Figure 1 shows the resulting diffractograms for graphite, graphite oxide, and rGO.

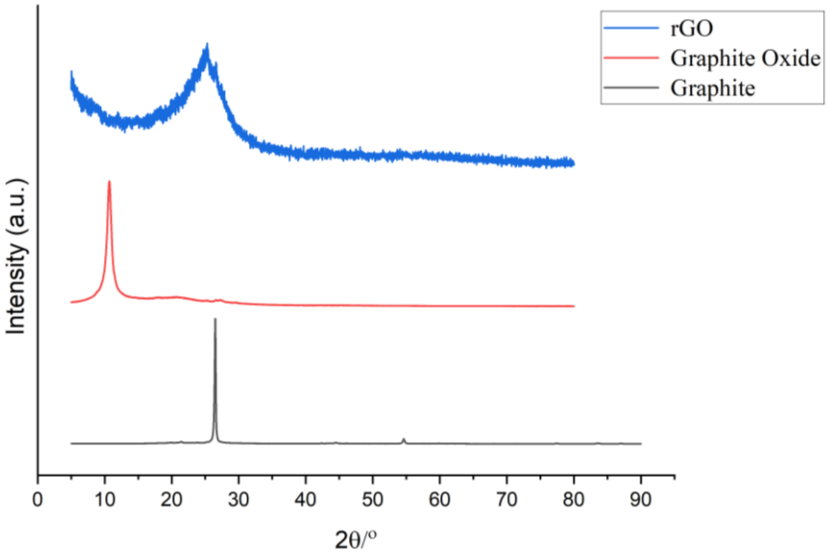


Figure 1. X-ray Diffractogram for Graphite, Graphite Oxide, and rGO

Graphite displayed values of 2θ = 26.4853° and d-spacing = 3.3626 Å with 99% crystallinity, this result is obtained by calculating the peak area divided by the total area. Graphite oxide displayed 2θ = 10.6926° and d-spacing = 8.2672 Å with 48.78%. The leftwards shift of 2θ to a lower value indicated a material change with fewer crystalline properties exhibited by the graphite oxide. The widening of d-spacing could be attributed to the formation of phenol, epoxy, ketone, carboxyl, and carbonyl groups that would facilitate exfoliation during the reduction process. This result is consistent with those reported in [6], [11], [15-17], where the average 2θ for graphite was 26° with a d-spacing of 3 Å while graphite oxide exhibited 2θ = 10° with a d-spacing of 8 Å. Finally, rGO displayed values of 2θ= 25.85° and d-spacing = 3.444 Å with 48.76% crystallinity. The rightwards shift indicates successful reduction of the graphite oxide, whereby the transformation from GO to rGO involved the loss of some phenol, epoxy, ketone, carbonyl, and carboxyl groups [13], [18]. This is supported by 19, which found rGO to have a 2θ value of 25°with a d-spacing of 3Å.

Fourier Transform Infrared (FTIR) Spectroscopy Analysis

FTIR analysis was performed to determine the presence of functional groups from the spectrum of vibrations observed in the sample. Figure 2 shows the FTIR spectra for graphite, graphite oxide, and rGO.

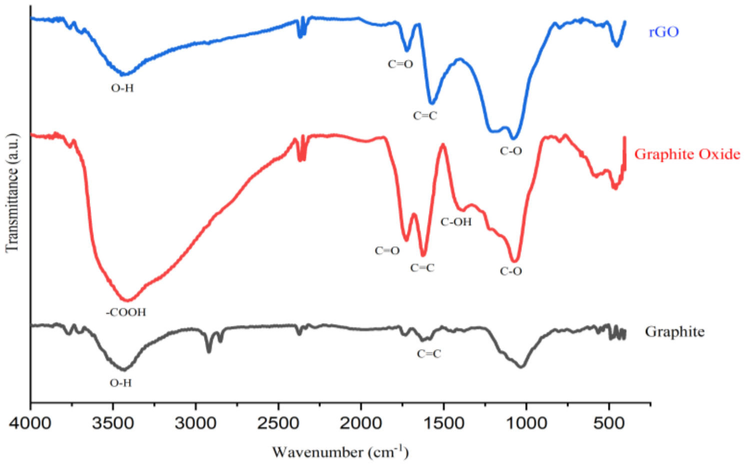
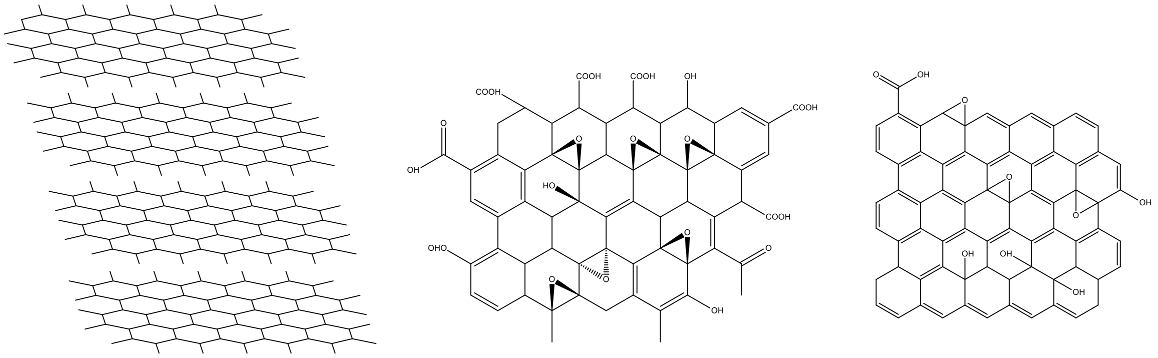


Figure 2. FTIR Spectra for Graphite, Graphite Oxide, and rGO

According to the FTIR spectrum, the graphite sample exhibited O-H vibrations at a wavelength of 3429.43 cm^{-1}, C-O at a wavelength of 1737.86 cm^{-1}, and C=C at 1583.56 cm^{-1}. This result is in accordance with those reported in [18]. The graphite oxide spectrum exhibited a stretching of O-H vibrations for hydroxyl groups at a wavelength of 3412.08 cm^{-1}, stretching of C=O vibrations for carboxyl groups at a wavelength of 1724.36 cm^{-1}, C=C vibrations at a wavelength of 1627,92 cm^{-1}, and the appearance of a new intensity for C=O vibrations (1737.86 cm^{-1}), C-OH vibrations (1381.03 cm^{-1}), and C-O vibrations (1074,35 cm^{-1}), in accordance with the results in [16], [18], [20]. These results indicated the successful synthesis of graphite oxide by the formation of oxygen-based functional groups such as -COOH, C=O, C=C, C-OH, and C-O. Graphite oxide is a lattice of sp^2 and sp^3 carbon atoms with insulating properties [5], and the results of this oxidation from graphite into graphite oxide should facilitate the exfoliation process during the reduction of the graphene oxide [18], [21]. Finally, rGO exhibited the partial or total loss of the oxygen group C-OH, which indicated successful reduction by the presence of O-H vibrations at a wavelength of 3431.36 cm^{-1}, C=O (1722,43 cm^{-1}), C=C (1573.91 cm^{-1}), and C-O (1080,14 cm^{-1}). These results are in accordance with [18]. Structure Graphite, Graphite Oxide, and rGO are presented in Figure 3.



Graphite Graphite Oxide rGO

Figure 3. Structure Graphite, Graphite Oxide, and rGO [22].

Scanning Electron Microscope (SEM) Analysis

The rGO morphology was analyzed with an SEM. The SEM observation results of the rGO samples at a magnification 10000x are presented in Figure 4.

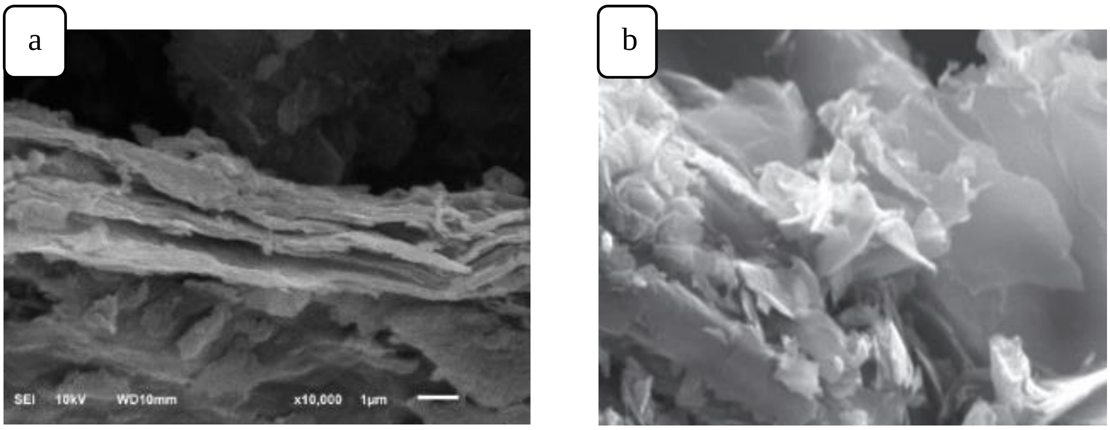


Figure 4. SEM microgram of a). rGO syntheized in this study b). rGO synthesized in [23].

According to figure 4, the morphology of rGO at 10000x exhibited triple-layered foliation. This contrasts to the result in [23], where the sheets/layers appeared to be thicker with wrinkled, folded, and/or stacked layers. The triple-layered morphology of rGO in this study may be attributable to the residual presence of oxygen-based functional groups, which led to the development of separate thin layers. This conclusion is supported by the FTIR spectrum, which indicated the presence of functional groups in the form of O-H, C=C, C=O, and C-O.

Electric Conductivity and Production in rGO

The effectiveness of rGO as an electrical conductor can be affected by the degree of success achieved in the graphene oxide reduction process. The reduction method used in this study was based upon that described in [11]. Measurements of the rGO conductivity were made with an LCR meter – an instrument used to measure the inductance, capacitance, and resistance of components within an electrical circuit [24]. While electrical power generation/storage was measured with a digital multimeter [3]. These measurements produced a conductivity value for rGO of 0.000357005 S/c, a voltage of 1.21 Volts, and a current of 3.55 mA.

The solid-state battery electrolyte synthesized in this study exhibited a lower conductivity than those in [1-3], but achieved a higher voltage than [2]. An earlier study by [25]. achieved a conductivity of 6.945 x 10^{-6} S/cm in rGO from coconut shell charcoal and 1.806 x 10^{-5} S/cm. The synthesized rGO still has low conductivity, as this rGO contains defect structures due to the presence of O_2 insertion. This is supported by FTIR characterization, which shows the abundance of O_2 related functional groups such as O-H, C=C, C=O, and C-O. As a result, the charge carrier mobility is hindered and the arrangement of the atoms formed is less orderly, which disrupts electron transport.

CONCLUSION

The synthesis of rGO from graphite through the Hummers method and the reduction of graphene oxide has been successfully achieved, whereby the XRD characterization of the rGO produced a 2θ value of 25.85° with a d-spacing of 3.444 Å and 48.76% crystallinity, while FTIR characterization displayed vibrations from O-H groups at a wavelength of 3431.6 cm^{-1}, C=O (1722,43 cm^{-1}), C=C (1573,91 cm^{-1}), and C-O (1080,14 cm^{-1}), and SEM analysis at 10000x magnification displayed a triple-layered morphology. The rGO thus synthesized had a conductivity of 0.000357005 S/cm, and produced a voltage of 1.21 Volts and a current of 3.55 mA as a battery electrolyte.

REFERENCES

1 Yuningsih, L. M., Dikdik, M., & Hera, A.S. 2018. Effect of Swelling and Sonication Methods on Conductivity of Polyaniline-Cellulose from Corn Cob (Zea mays L). *International Journal of Applied Chemistry*. *14*(4), 281 – 292.

2 Yuningsih, L. M., Dikdik, M., & Yusri, M. F. 2018. Synthesis of Polyaniline-Activated Carbon Composite from Corn Cob as Solid Electrolyte in Batteries. *Journal of Chemistry Valence*. *4*(2), 119 - 123.

3 Yuningsih, L. M., Dikdik, M., & Yusri, M. F. 2021. Synthesis of Polyaniline Microcellulose Composite. *Journal of Chemistry and Packaging*. *43*(2), 50 – 55.

4 [BSN] National Standarization Agency of Indonesia. 2004. SNI 04.2051.2.2004. Primary Battery Part 2: Physical and Electrical Spesification. National Standarization Agency of Indonesia, Jakarta.

5 Dwandaru, W. S. B., Rhyko, I. W. W., & Lia, D. P. 2019. *Nanomaterials Graphene Oxide synthesis and characterization*. UNY Press, Yogyakarta.

6 Suwandana, R. F., & Diah, S. 2015. Analysis of the influence of Zn reductan mass on the capacitive properties of graphene material supercapacitor. *Journal Technique ITS*. 4. F95 – F100.

7 Wang, C., Jianwei, Z., & Feipeng, D. 2016. Synthesis Highly Reduced Graphene Oxide for Supercapacitor. *Journal of Nanomaterials.* 1 – 7.

8 Patel, D. K., Yupri, S., & Ki-Taek, L. 2019. Stimuli-Responsive Graphene Nanohybrids for Biomedical Applications. *Journal Stem Cells International*. 1 – 18.

9 Tao, Z., Jianrong, X., Zhiyun, Y., & Heng, W. 2019. Graphene/Sulfur @Graphene Composite Structure Material for a Lithium-sulfur Battery Cathode. *Journal of Nanomaterial*. 1 – 10.

10 Mann, R., Dimotrios, M., Zhirun, X., Oliver, M., Yun, H. N., Rose, A., & Cindy, G. 2021. Antibacterial Activity of Reduced Graphene Oxide. *Journal of Nanomaterial*. 1 – 10.

11 Taufantri, Y., Irdawati., Raka, I. A., & Asih, A. 2016. Synthesis and Characterization of Graphene with Graphite Oxide Reduction Method Using Zn Reducer. *Journal of Chemistry Valence*. *2*(1), 17 – 23.

12 Narasimharao, K., Venkata, R. G., Sreedhar, D., & Vasudevaro, V. 2016. Synthesis of Graphene Oxide by Modified Hummers Method and Hydrotermal Synthesis of Graphene NiO Nano Composite for Supercapacitor Application. *Journal of Material Sciences and Engineering*. *5*(6), 1 -4.

13 Chen, X., Zhan, Q., Zhe, L., & Guoyu, R. 2022. Mechanism of Oxidation of Graphite to Graphene Oxide by the Hummers Method. *Journal ACS OMEGA*. 7. 235603 – 23510.

14 Anwar, D. I., Lela, L. K., Nurbaeti, N., & Eri, R. H. 2023. Synthesis of Reduced Graphene Oxide Based on Rubber Seed Shells and Rice Husks and Their Composites Using the Modified Hummer Method.*: Erath and Enviromental Science*. 1-7.

15 Cao, N., & Yuan, Z. 2015. Study of Reduced Graphene Oxide Preparation by Hummers' Method and Related Characterization. *Journal of Nanomaterials*. 1 -5.

16 Habte, A. T., & Delele, W. A. 2019. Synthesis and Characterization of Reduced Graphene Oxide (rGO) Started from Graphene Oxide (GO) Using the Tour Method with Different Parameters. *Advances in Materials Science and Engineering*. 1 – 9.

17 Syukri, S., Fadhil, F., Yetria, R., Yulia, E. K., Mai, E., & Upita, S. 2021. Synthesis of Graphene Oxide Enriched Natural Kaolinite Clay and Its Applications for Biodiesel Production. *Journal of Renewable Energy Depelopment*. *10*(2), 307-315.

18 Rochman, R. A., Sayekti, W., Ari, H. R., & Qonita, A. W. 2019. Preparation of Nitrogen and Sulphur Co-doped Reduced Graphene Oxide (rGO-Ns) Using N and S Heteroataom of Thiourea.

IOP Conference Series: *Materials Science and Engineering*. IOP Publishing. Strankowski, M., Damian, W., Lukasz, P., & Justyna, S. 2016. Polyurethane Nanocomposites Containing Reduced Graphene Oxide, FTIR, Raman, and XRD Studies. *Journal of Spectroscopy*. 1 – 6.

19 Sujiono, E. H., Zumansyah., Zabrian, D., Dahlan, M. Y., Amin, B. D., Samnur., & Agus, J. 2020. Graphene Oxide Based Coconut Shell Waste: Synthesis by Modified Hummers Method and characterization. *Journal Heliyon*. 1-8.

20 Faniyi, I. O., Fasikin, O., Olofinjana, B., Adekunle, A. S., Oluwasusi, T. V., Eleruja, M. A., & Ajayi E. O. B. 2019. The comporative analyses of reduced grapheme oxide (rGO) prepared via green, mild and chemical approaches. *Journal Springer Nature*. 1 -7.

21 Naraprawatphong R, Chayanaphat Chokradjaroen, Satita T, Li Y, & Nagahiro S. 2022. Nanoscale Advanced Carbons as an Anode for Lithium-ion Btterey. *Journal Materials Today Advances*. 1-30.

22 Syalaby, A., Nihtianova, D., Markov, P., Staneva, A.D., Iordanava, R.S., & Dimitriev, Y.B. 2015. Structural analysis of reduced graphene oxide by transmission electron microscopy. *Bulgarian Chemical Communications*. *47*(1), 291-295.

23 Simic, M. 2014. Realization of Digital LCR Meter. *International Conference and Exposition on Electrical and Power Engineering (EPE 2014)*. 769 – 773.

24 Putri, N.A., Hikmah, U., Prasetyo, A. 2023. Green Synthesis of Reduced Graphene Oxide from Coconut Shell Charcoal and Wood Using Enviromentals Reductor L-Ascorbic Acid. *Journal Chemistry*. *17*(1), 82-88.