

SYNTHESIS OF GRAPHENE LIKE CARBON (GLC) FROM COCONUT SHELLS WITH HIGH AND STABLE ELECTRICAL CONDUCTIVITY

NUR ROHMAT*, #JON AFFI**, GUNAWARMAN GUNAWARMAN**, MURNI HANDAYANI***, SYAIFUL BAKHRI*, YULI YETRI****, GIYANTO*

*Mechanical Engineering Dept, Pamulang University, Surya Kencana st. No.1,
South Tangerang 15417, Indonesia

**Mechanical Engineering Dept., Andalas University, Kampus Limau Manis, Padang 25163, Indonesia

***Research Center for Advanced Materials, National Research and Innovation Agency
(BRIN), Indonesia

****Mechanical Engineering Dept., State Polytechnic of Padang, Padang 25163, Indonesia

#E-mail: jon_affi@eng.unand.ac.id

Submitted November 12, 2024, accepted January 28, 2025

Keywords: Graphene, XRD, SEM-EDX, Raman, FTIR, Electrical Conductivity

This research focuses on synthesising, the characterisation, and electrical conductivity testing of graphene nanosheets made from the renewable resource of coconut shells. Graphene and similar carbon-based nanomaterials, with their unique physical, chemical, and electrical properties, are ideal for battery cathode applications. The synthesis involves roasting whole old coconut shells at 150 °C and 200 °C, followed by pyrolysis at 750 °C to prepare graphene-like carbon. The synthesised material was characterised using XRD, SEM-EDS, RAMAN, FT-IR, and electrical conductivity tests. The XRD results indicated the presence of graphene peaks and oxygen-containing graphene functional groups. The SEM results revealed typical graphene surface morphology with interlocking layers of porous three-dimensional graphene. The EDX analysis confirmed carbon as the majority compound content, with a minor presence of oxygen, validating the synthesis of graphene. The Raman spectroscopy showed similar intensities at different temperatures, indicating the formation of more SP² domains. The FT-IR testing confirmed the bonding interaction of graphene at both temperatures, demonstrating the presence of carbon groups. The electrical conductivity tests show that the obtained graphene nanosheet has high and stable conductivity, which can effectively control electron mobility by storing.

INTRODUCTION

Graphene is one of the most studied materials to this day. The basic properties of graphene make it very promising in some applications, such as electronics, mechanics, and optics. Notably, graphene sheets have a theoretical specific surface area of 2630 m²·g⁻¹, which has attracted great interest in energy storage applications, including supercapacitors and batteries [1].

Today, manufacturers of supercapacitors, electrodes, fuel cells, supercapacitors, cathode materials for Li-ion capacitors, and batteries have a wide range of electrochemical potential. Most of the applications use activated carbon made from coconut shells as the active ingredient in their supercapacitor electrodes due to their high specific surface area and low price, making them capable of mass production [1, 2, 3]. Coconut shells account for about 85 % of the fruit's weight [4], having an assumed composition of 33.30 % lignin, 30.58 % cellulose, 26.70 % hemicellulose, 8.86 % water and 0.56 % ash [5], [6], which holds promise as a potential

source of energy from the raw materials [7]. These shells can also be transformed into graphite and graphene nanosheets [8], further enhancing their value in our research. Recent research has revealed that graphene, a product of coconut shells, can significantly enhance the activity of catalysts and the chemical interactions of electrodes, fuel cells, and primary batteries [9], [10]. This exciting finding underscores the potential of renewable graphene in advancing the field of energy science.

Developing advanced materials for renewable energy increasingly involves the use of graphene nanosheets due to their unique properties, though they present both benefits and challenges. Graphene offers several key advantages: (a) it improves the electrical performance and provides excellent chemical stability when used in batteries [11]; (b) its specific surface area ranges from about 100 to 1500 m²·g⁻¹ and can theoretically reach as high as 2630 m²·g⁻¹ [12]; (c) it serves as an effective support material for alloyed metal particles [13]; (d) graphene exhibits high electrical and

thermal conductivity [14]; (e) its structure is extremely thin, flexible, and remains stable under both chemical and thermal conditions [11]; (f) graphene's π -conjugate bonds and high electron mobility further enhance its electrical properties [15]. This combination of properties makes graphene a valuable material in the development of renewable energy technologies, though careful consideration of its limitations is also essential.

Previous studies have advanced the use of graphene in primary battery cathodes, showing promising results for practical applications [16]. The integration of graphene into the cathode material can improve the electron conductivity, enhancing the overall electrical conductivity of the cathode. Additionally, incorporating metal into this set-up can increase the cathode activity and improve the graphene electronic interaction quality [17]. These advantages highlight the real-world potential of graphene in battery technology. This research focuses on several key concepts: synthesising graphene-like carbon (GLC) from natural sources, such as coconut shells; exploring graphene's potential to increase the energy storage capacity; and assessing its ability to enhance the specific energy and energy capacity in batteries. Graphene nanosheets are particularly suitable for primary battery cathodes [18], and the synthesis process is relatively simple. These findings emphasise the value of graphene as a versatile material in developing advanced energy storage solutions.

EXPERIMENTAL

This study obtained the graphene material from the shell of aged coconuts. Initially, the coconuts were dried at temperatures of 150 °C and 200 °C in an aluminium kettle. The coconut shell was then cleaned and heated at 300 °C for an hour. Following this, the temperature was raised to 750 °C to make charcoal. The produced charcoal was ground and sieved to a 200-mesh size. Next, the charcoal powder was held in distilled water for 24 hours, followed by filtration using Whatman paper and drying at room temperature for another 24 hours. This was further processed by sonication for one hour and then dried again at room temperature for 24 hours. A final drying step was conducted on a hotplate at 100 °C for 4.5 hours. The samples were subsequently characterised using techniques such as X-ray diffraction (XRD), Scanning electron microscopy-Energy-dispersive X-ray spectroscopy (SEM-EDX), Raman spectroscopy, and Fourier transform infrared (FTIR) spectroscopy. Two specific samples are documented: D1, which is dried at 150 °C, and D2, dried at 200 °C, both subjected to initial combustion at 300 °C for one hour and a consistent pyrolysis temperature of 750 °C for an hour [19]. The electrical conductivity is determined through a test using a compacted fuse weighing about 0.40 grams, covered with a fuse cover. The clamp wires

are connected to a digital multimeter's negative and positive poles. The electrical conductivity is measured with voltage variations of 40, 45, 50, and 55 volts with a current of 9.4 Amperes [9]. Other tools used for this conductivity test include a power supply, which supplies the electric current. The conductivity test calculation can be determined using $R = V / I$ and $DHL = 1 / R$ [10].

RESULT AND DISCUSSION

Graphene-like Carbon Characterisation

In order to derive the graphene-like carbon (GLC) from old coconut shell materials, two experiments were carried out with different initial drying temperatures: D1 drying at 150 °C and D2 drying at 200 °C, both with a pyrolysis of 750 °C. The resulting charcoal powder from the old coconut shell was then characterised using an X-ray diffraction analysis. This technique measures the average distance between the layers or rows of atoms, as shown in Figure 1. It is known that the diffraction peak of pure graphite is at about 2θ at 26.5° and 42.5° with an interlayer distance of 0.335 nm [20, 21]. In this case, graphene with D1 shows diffraction peaks at 2θ of 24.230 with a distance of 0, 371 nm and 43.410 with a distance of 0, 205 nm, typical graphene peaks. Similarly, D2 shows diffraction peaks at 2θ of 23.820 with a distance of 0, 206 nm and 43.410 with a distance of 0, 205 nm. The diffraction peak of the sample corresponds to the reference [20, 21].

The observations from the Raman spectroscopy are depicted in Figure 2. The G band, characteristic of all graphite structures, arises due to the bond stretching motion in the sp^2 plane of the hybridised carbon atoms. The D and G values of D1 at 1336.52 cm^{-1} and 1590.11 cm^{-1} , respectively, and at D2 are 1337.69 cm^{-1}

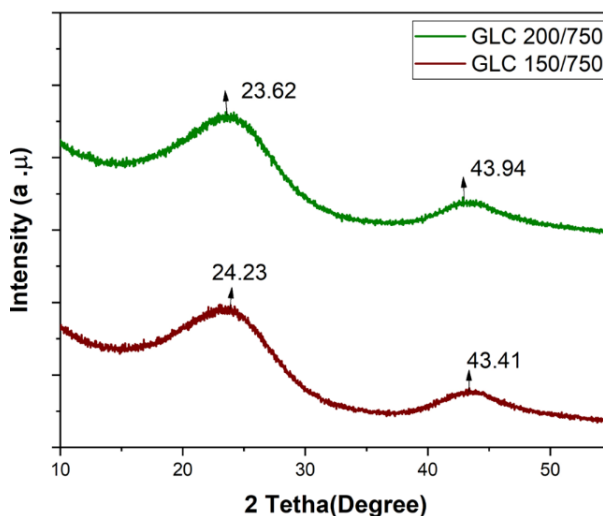


Figure 1. Characterisation results of the GLC XRD at the initial D1 and D2.

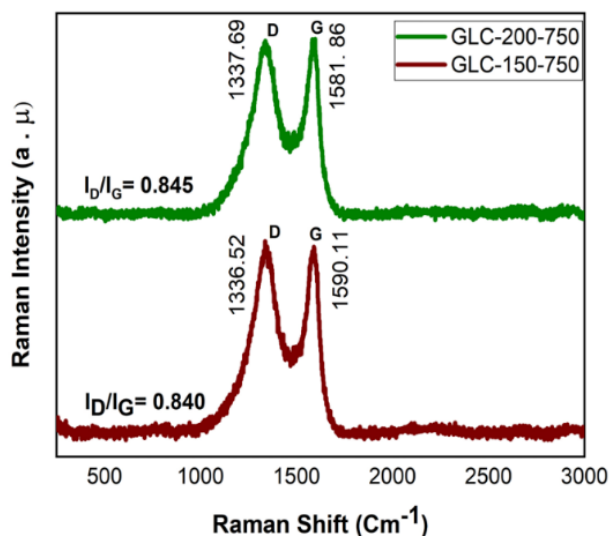


Figure 2. Characterisation results of the GLC Raman at the initial D1 and D2.

and 1581.86 cm^{-1} . The Raman intensities produced at different temperatures are almost identical. The I_D/I_G of D1 is 0.840, and D2 is 0.845, indicating that more sp^2 domains are formed and there are no defects in the graphene with the increasing pyrolysis temperature [20].

The FT-IR technique is used to confirm the bonding interactions of graphene, as shown in Figure 3. For D1, the OH stretching vibrations indicate the strong and wide band at 3444.5 cm^{-1} . The peak in the range 2863.42 cm^{-1} to 2375.02 cm^{-1} indicates a C = C bond. The peak in the ring of 1541 , 34 and 1121.82 cm^{-1} is attributed to the C = O stretching vibration and the peak at $876, 55 \text{ cm}^{-1}$ is attributed to the COC bonding, which agrees with previous reports [22]. For D2, the OH stretching vibrations indicate the strong and wide band at 3430.69 cm^{-1} . The peak is in the range of 2928.81 cm^{-1} to 2028.30 cm^{-1} indicates a C = C bond. The peak in the

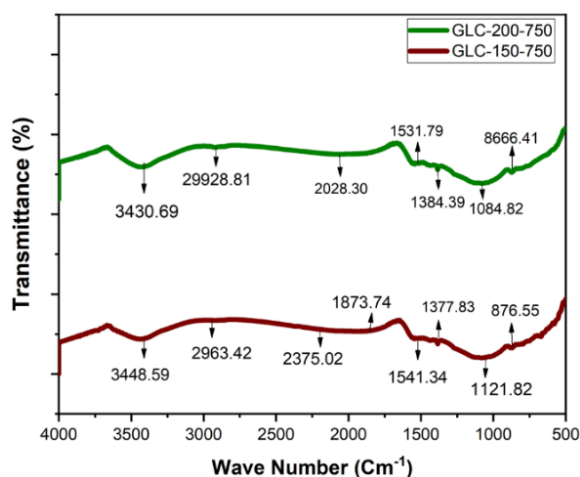


Figure 3. Characterisation results of the GLC FT-IR at initial the D1 and D2.

range of 1531.79 cm^{-1} to 1384.39 cm^{-1} indicates a C = C bond. The peak at 1084.82 cm^{-1} is attributed to the C = O stretching vibration, and the peak at 8666.41 cm^{-1} is attributed to COC.

The detailed morphology characterisation revealed slightly thick edges, not centred, with the image magnified 50,000 times. The synthesis results demonstrated that the typical surface morphology of graphene was observed, with layers interlocking to form the porous three-dimensional graphene structure seen in 4(a). The EDX analysis, as shown in Figure 4(b), provided intriguing results. It indicated that most of the compound content was carbon (96 % by mass), followed by oxygen (4 % by mass), confirming the presence of graphene; the presence of oxygen (4 % by mass) suggested that the sample's neutralisation was successful, raising interesting questions for further investigation. For sample D2, shown in Figure 5(a), the edges were slightly thick and not centred, with the image magnified 50,000 times. The EDX analysis result, shown in Figure 5(b), revealed that the elemental composition of GLC was 93.2 % carbon (C) and 6.8 % oxygen (O). Ideally, GLC should only contain carbon in its composition. However, oxygen was found, indicating a defect caused by the amorphous synthesis process during pyrolysis.

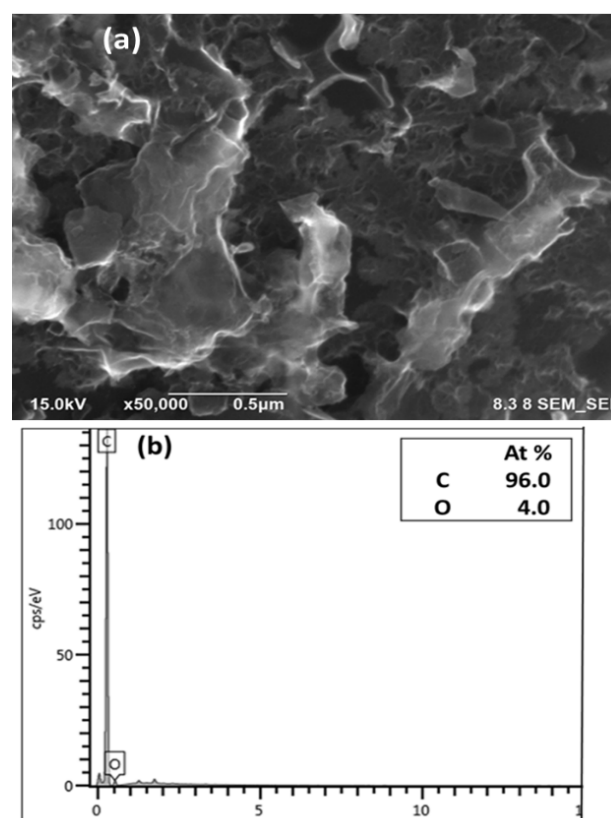


Figure 4. (a) Presents our meticulous Scanning Electron Microscopy (SEM) – (b) EDX analysis on the surface of the GLC at the initial D1.

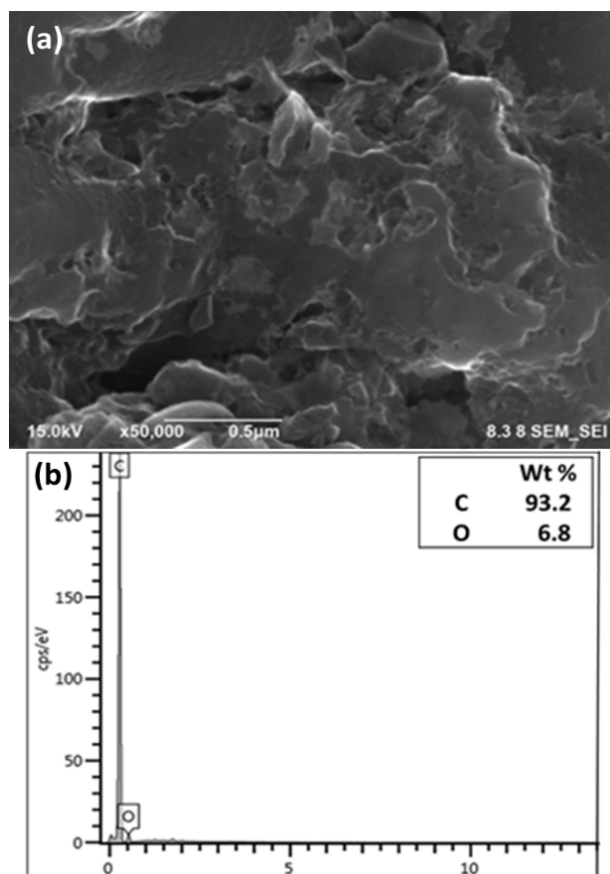


Figure 5. The meticulous Scanning Electron Microscopy (SEM) - EDX analysis on the surface of GLC at the initial D2.

Electrical conductivity test of the GLC

TA conductivity test was conducted to assess the stability of GLC's electrical conductivity. The test data, which can be viewed in Figure 6, revealed that, at D1, the electrical conductivity was $1.686 \mu\text{S}\cdot\text{cm}^{-1}$ at 40 V and $1.111 \mu\text{S}\cdot\text{cm}^{-1}$ at 55 V. For D2, the electrical conductivity was $1.519 \mu\text{S}\cdot\text{cm}^{-1}$ at 40 V and $1.087 \mu\text{S}\cdot\text{cm}^{-1}$ at 55 V. It was observed that as the voltage increased, the electrical conductivity decreased.

The electrical conductivity of GLC demonstrated a relatively stable decrease, losing electrons intermittently with a minor increase in the current. This characteristic indicates that it allows for the better control over the electron mobility. It is probably caused by the ability of graphene to store electrons and release them slowly. However, as the pyrolysis temperature increases, the electrical conductivity of graphene decreases [23]. The stability of graphene's conductivity, a factor of paramount importance, plays a crucial role in conducting electric current and in the lifespan of graphene-based batteries.

CONCLUSION

Graphene-like carbon (GLC) was successfully synthesised from coconut shells through heat at varying drying temperatures and ultrasonic procedures. The presence of GLC can be confirmed through various characterisation techniques such as XRD, SEM-EDX, Raman spectroscopy, and electrical conductivity tests. The GLC obtained from this process can be used as a battery electrode material. The XRD results, a key indicator of successful graphene synthesis, unequivocally showed the presence of graphene peaks and the formation of oxygen-containing graphene functional groups. The SEM tests, a reliable method for surface analysis, revealed the typical surface morphology of graphene, characterised by interlocking layers of porous three-dimensional graphene. The elemental analysis conducted through an EDX analysis, a standard technique for identifying elements in a sample, showed that most of the compound content is carbon, with a small amount of oxygen, confirming the presence of graphene.

Observations from Raman spectroscopy showed that the Raman intensity produced at different temperatures did not significantly differ. The result can be observed in the nearly identical ID/IG intensity ratio, indicating that increasing the drying temperature does not significantly influence the results. The FT-IR testing confirmed that the GLC possesses several oxygen and carbon functional groups. The electrical conductivity tests on samples D1 and D2 demonstrated that the GLC can effectively control the electron mobility with high and stable conductivity. This finding has significant implications for developing high-performance battery electrodes, where controlled electron mobility is crucial.

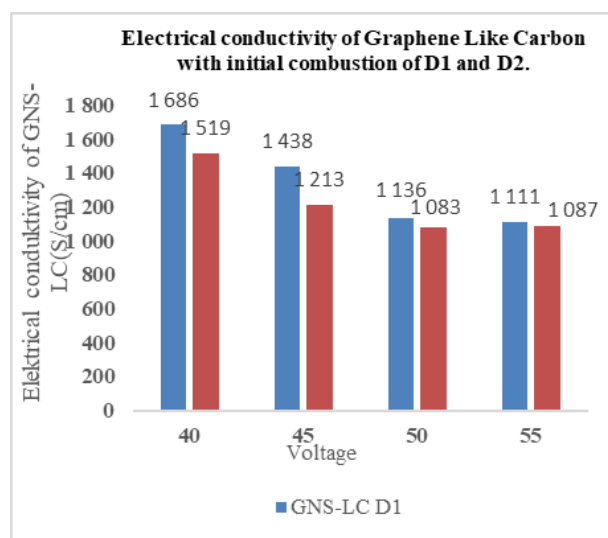


Figure 6. Electrical conductivity of GLC for D1 and D2.

ACKNOWLEDGMENT

The authors express their gratitude to all those who participated in completing this research. This work received support from the Directorate of Higher Education in Indonesia, facilitated through Andalas University, which initiated a competitive national research programme. This programme specifically catered to doctoral research dissertations, with the contact number being 86/UN16.19/PT.01.03/2023. The authors would also like to extend special thanks to the National Research and Innovation Agency (BRIN) for allowing the characterisation material devices to be utilised.

REFERENCES

- Gao Y. (2017): Graphene and polymer composites for supercapacitor applications: a review. *Nanoscale research letters*, 12(1), 387. doi: 10.1186/s11671-017-2150-5
- Jain A., Aravindan V., Jayaraman S., Kumar P. S., Balasubramanian R., et al. (2013): Activated carbons derived from coconut shells as high energy density cathode material for Li-ion capacitors. *Scientific reports*, 3(1), 3002. doi: 10.1038/srep03002
- Ganguly S., Sengupta J. (2024): Graphene-based nanotechnology in the Internet of Things: a mini review. *Discover Nano*, 19(1), 110. doi: 10.1186/s11671-024-04054-0
- Ayrlimis N., Jarusombuti S., Fueangvivat V., Bauchongkol P., White R. H. (2011): Coir fiber reinforced polypropylene composite panel for automotive interior applications. *Fibers and polymers*, 12, 919-926. doi: 10.1007/s12221-011-0919-1
- Arena N., Lee J., Clift R. (2016): Life Cycle Assessment of activated carbon production from coconut shells. *Journal of Cleaner Production*, 125, 68-77. doi: 10.1016/j.jclepro.2016.03.073
- Nunes L. A., Silva M. L., Gerber J. Z., Kalid R. D. A. (2020): Waste green coconut shells: Diagnosis of the disposal and applications for use in other products. *Journal of Cleaner Production*, 255, 120169. doi: 10.1016/j.jclepro.2020.120169
- Sarkar J. K., Wang Q. (2020): Different pyrolysis process conditions of South Asian waste coconut shell and characterization of gas, bio-char, and bio-oil. *Energies*, 13(8), 1970. doi: 10.3390/en13081970
- Supeno M., Siburian R. (2020): New route: Conversion of coconut shell to be graphite and graphene nano sheets. *Journal of King Saud University-Science*, 32(1), 189-190. doi: 10.1016/j.jksus.2018.04.016
- Wasalathilake K. C., Li H., Xu L., Yan C. (2020): Recent advances in graphene based materials as anode materials in sodium-ion batteries. *Journal of Energy Chemistry*, 42, 91-107. doi: 10.1016/j.jechem.2019.06.016
- Simanjuntak C., Siburian R., Marpaung H. (2020): Properties of Mg/graphite and Mg/graphene as cathode electrode on primary cell battery. *Heliyon*, 6(1), e03118. doi: 10.1016/j.heliyon.2019.e03118
- Zhu J., Duan R., Zhang S., Jiang N., Zhang Y., Zhu J. (2014): The Application of Graphene in Lithium Ion, *Springer Plus*, 585, 1-8.
- Benti N. E., Tiruye G. A., Mekonnen Y. S. (2020): Boron and pyridinic nitrogen-doped graphene as potential catalysts for rechargeable non-aqueous sodium-air batteries. *RSC advances*, 10(36), 21387-21398. doi: 10.1039/d0ra03126g
- Ali A., Shen P. K. (2019): Recent advances in graphene-based platinum and palladium electrocatalysts for the methanol oxidation reaction. *Journal of Materials Chemistry A*, 7(39), 22189-22217. doi: 10.1039/c9ta06088j
- Yaqoob A. A., Ibrahim M. N. M., Yaakop A. S., Umar K., Ahmad A. (2021): Modified graphene oxide anode: a bioinspired waste material for bioremediation of Pb²⁺ with energy generation through microbial fuel cells. *Chemical Engineering Journal*, 417, 128052. doi: 10.1016/j.cej.2020.128052
- Samuels A. J., Carey J. D. (2013): Molecular doping and band-gap opening of bilayer graphene. *Acs Nano*, 7(3), 2790-2799. doi: 10.1021/nn400340q
- Guo Q., Zeng W., Liu S. L., Li Y. Q., Xu J. Y., Wang J. X., Wang Y. (2021): Recent developments on anode materials for magnesium-ion batteries: a review. *Rare Metals*, 40(2), 290-308. doi: 10.1007/s12598-020-01493-3
- Guo Q., Zeng W., Liu S. L., Li Y. Q., Xu J. Y., Wang J. X., Wang Y. (2021): Recent developments on anode materials for magnesium-ion batteries: a review. *Rare Metals*, 40(2), 290-308. doi: 10.1007/s12598-020-01493-3
- Vu T. T., Eom G. H., Lee J., Park M. S., Moon J. (2021): Electrolyte interface design for regulating Li dendrite growth in rechargeable Li-metal batteries: A theoretical study. *Journal of Power Sources*, 496, 229791. doi: 10.1016/j.jpowsour.2021.229791
- Giyanto J. A., Gunawarman, Handayani, Yetri Y., Rohmat N. (2024): Characterisation M. of Graphene Derived From Coconut Shells: Impact of Ammonia Doping and the Sonication Method. *Ceramics - Silikaty*, 68, 116-120. doi: 10.13168/cs.2024.0010
- Maulana A., Nugraheni A. Y., Jayanti D. N., Mustofa S., Baqiya M. A. (2017): Defect and magnetic properties of reduced graphene oxide prepared from old coconut shell. In *IOP Conference Series: Materials Science and Engineering* (Vol. 196, No. 1, p. 012021). IOP Publishing. doi: 10.1088/1757-899X/196/1/012021
- Siburian R., Sihotang H., Raja S. L., Supeno M., Simanjuntak C. (2018): New route to synthesise of graphene nano sheets. *Oriental Journal of Chemistry*, 34(1), 182. doi: 10.13005/ojc/340120
- Prasetya F. A., Nasrullah M., Nugraheni A. Y., Darminto D. (2015). Study of Raman spectroscopy on graphene phase from heat treatment of coconut (Cocos Nucifera) shell. In *Materials Science Forum* (Vol. 827, pp. 290-293). Trans Tech Publications Ltd. doi: 10.4028/www.scientific.net/MSF.827.290
- Hutagalung F. Y. S. T., Siburian R., Supeno M. (2021): The performance conductivity of Mg/Graphene nanosheet as anode of battery. In *IOP Conference Series: Materials Science and Engineering* (Vol. 1122, No. 1, p. 012090). IOP Publishing. doi: 10.1088/1757-899X/1122/1/012090