Design and Performance Optimisation of Graphite-Based Batteries for Advanced Energy Storage System

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Abstract

This research focuses on the development of a graphite-based battery, utilising the abundant graphite resources available in Sri Lanka while facing the challenges of limited access to advanced materials and technologies. The primary objective is to create a viable graphite battery that can potentially contribute to the advancement of energy storage technologies. Given the constraints in accessing certain materials and technologies, this study explores the electrochemical properties of graphite and investigates its potential application as a key component in battery design. Through systematic testing and optimisation processes, we aim to enhance the battery's performance and energy storage capacity. The outcomes of this research provide valuable insights into the potential of graphite-based batteries, serving as a stepping stone for future improvements and innovations in energy storage. Although the developed battery may require further enhancements, it contributes to the growing body of knowledge in sustainable energy solutions, benefiting both Sri Lanka and the global energy sector.

Keywords: Activated carbon, Aluminum, Electrode, Electrolyte, Expanded graphite

1 Introduction

Batteries are devices that store electrical energy and convert it into chemical energy, which can then be used to power devices and machines [1]. They are essential components in many everyday devices, including smartphones, laptops, and electric vehicles. However, there are several shortcomings of batteries such as reliability, efficiency, cost, etc. [2] that have led researchers to search for new materials and technologies to improve their performance.

There are different types of batteries available on the market today, including lithium-ion batteries, lead-acid batteries, and others [3]. One of the main shortcomings of lithium-ion batteries is their relatively expensive compared to other types of batteries [4]. They also require special handling and disposal due to the presence of toxic materials. Lead-acid batteries, while less expensive, also have a shorter lifespan and can be heavy and bulky [5].

However, in Sri Lanka, there are additional challenges in producing batteries due to import restrictions and a lack of materials. This can make it difficult for manufacturers to access the necessary materials and technology to produce high-quality batteries.

One solution to these problems is the use of graphite-based batteries, which have been the subject of much research in recent years [6]. Graphite is a form of carbon that is abundant, inexpensive, and easy to extract, making it an attractive material for battery production.

Graphite-based batteries have garnered attention as a potentially cost-effective traditional alternative to lithium-ion batteries. In recent years, the demand for energy storage solutions has increased significantly due to the proliferation of renewable energy sources and the need for stable and reliable power grids. In this context, graphite-based batteries have the potential to play a significant role as they can offer high energy density, long cycle life, and fast charging capabilities at a lower cost compared to traditional lithium-ion batteries [7].

Sri Lanka is well known for high-quality vein graphite, containing 95-99% of pure carbon [8]. Graphite is typically found in veins or lenses within metamorphic rocks and is often mined using traditional, manual methods.

Graphite's high theoretical specific capacity allows it to store more ions per weight [9], leading to better energy density and battery life. Also, graphite is highly conductive due to its weakly bonded carbon layers, making it a good choice for battery production [10]. This allows for electrons to move easily through graphite's weakly bonded layers [11]. Graphite also conducts heat well [12].

Despite these advantages, the adoption of graphite-based batteries has been limited due to a lack of understanding of their performance and degradation mechanisms. There is a need to understand how the microstructure of the graphite electrodes and the operating conditions influence battery performance [13].

This research can help develop cheaper and more reliable energy storage. In particular, the results of this study could be used to optimise the design of graphite-based batteries for specific applications and to identify strategies for improving their performance and stability.

2 Methodology

The methodology encompasses a series of steps, starting with the fabrication of graphite electrodes using a binder and conductive additive. Electrode drying and conditioning procedures are implemented to ensure the stability and uniformity of the electrode materials. The assembled battery cells undergo rigorous testing, including initial characterisation, cyclic voltammetry, and charge/discharge cycling.

Data analysis techniques are used to interpret results and draw conclusions during experimentation. Statistical analysis assesses the significance of differences or graphite-based trends and compares lithium-ion batteries batteries to or reference systems. Additionally, safety considerations and control experiments are implemented to ensure the reliability, reproducibility, and accuracy of the findings.

2.1 Developing expanded graphite

This research project uses expanded graphite as a crucial material for electrode fabrication. This material offers desirable properties for electrode applications, including large surface area, improved electrical conductivity, ion transport, and mechanical stability.

To obtain expanded graphite for electrode fabrication, a method involving the addition of hydrogen peroxide to graphite is employed. Specifically, a solution is prepared by adding 20 ml of hydrogen peroxide to 100 g of graphite. The hydrogen peroxide serves as an intercalant, facilitating the expansion of the graphite flakes during subsequent thermal treatment [14]. This intercalation process involves the incorporation of the hydrogen peroxide molecules between the layers of graphite, creating space for expansion upon heating [15].



Figure 1: Expanded graphite preparation process.

2.2 Electrode preparation

Thermal Treatment and Dewatering: The process begins by subjecting the expanded graphite to a controlled thermal treatment, typically in a hot plate, to remove any residual moisture present in the material. This step ensures the stability and integrity of the expanded graphite by eliminating moisture that could adversely affect electrode performance.

Mold Preparation and graphite loading: A specifically designed mold, tailored to the desired electrode dimensions with a diameter of 5 cm and height of 6 cm, is prepared for shaping the expanded graphite. The expanded graphite, now in a dry state, is carefully placed into the mold, ensuring uniform distribution and proper alignment within the mold cavity.

Hydraulic Pressing: To transform the loose expanded graphite into a compact and uniform electrode sheet, a hydraulic press is employed. The loaded mold, containing the expanded graphite, is positioned within the press. Applying controlled pressure, typically through a hydraulic system, the expanded graphite is compressed and compacted, resulting in the formation of a dense electrode sheet.



Figure 2: Electrode preparation process



Figure 3: Mold that used to create the electrodes.

2.3 Electrolyte preparation

For the battery electrolyte, a carefully formulated mixture comprising activated carbon and sulfuric acid (H_2SO_4) is utilised. Additionally, sodium sulfate (Na₂SO₄), bicarbonate phosphoric acid, sodium (NaHCO₃), and sodium chloride (NaCl) are incorporated into the electrolyte composition. This comprehensive blend of additives is strategically chosen to enhance the electrolyte's conductivity, pH stability, and overall electrochemical performance [16]. Activated carbon, known for its high surface area and porosity, aids in facilitating ion transport within the electrolyte, promoting efficient charge transfer processes [17]. The inclusion of sulfuric acid provides an acidic environment, which can enhance the ion mobility and improve the overall battery efficiency. Furthermore, the addition of Na₂SO₄, phosphoric acid, NaHCO₃, and NaCl assists in adjusting the electrolyte's composition, optimising its properties to meet the specific requirements of the graphite-based batteries in this research project.

2.4 Separators preparation

The separators consist of various materials. High porosity and filtering qualities of filter sheets enable effective ion transport without allowing for physical contact between the electrodes. Serviette paper offers good insulation and separator functioning when they have the right thickness and structural integrity. Furthermore, recycled lead-acid battery separators are used, leveraging their performance attributes proven and environmentally friendly nature. We hope to assess the effect of this wide variety of separator materials on battery performance, considering elements like ion diffusion, impedance, and overall electrochemical efficiency.

2.5 Current Collectors' preparation

Various current collector materials are used in this research effort, such as steel mesh, aluminum sheets, stainless steel sheets, and copper-coated PCB sheets. These materials are selected based on their effective current distribution, mechanical strength, electrical conductivity, resistance to corrosion, and lightweight nature. The goal is to look into how they affect the electrical efficiency, structural integrity, and stability of the battery system.

2.6 Battery testing

Several testing methodologies can be used to evaluate the performance and characteristics of the generated batteries. The following are some tests that can be performed on each battery configuration.

2.6.1 Capacity testing

Discharge the battery at a continuous rate and measure the time it takes for the voltage to decrease to a certain cutoff point to determine its capacity. The battery's energy storage capacity is determined by this test.

2.6.2 Voltage profile testing

Monitor the battery's voltage profile during charge and discharge cycles to determine the battery's voltage stability and consistency over time. This test assesses the battery's ability to maintain a consistent voltage output while in use.

2.6.3 Cycling testing

Charge and discharge the battery several times to evaluate its cycling performance and determine any capacity fading or degradation over time. This test reveals information about the battery's longevity and durability.

2.6.4 Self-discharge testing

Monitor the voltage decrease over a certain period when the battery is not in use to determine the self-discharge rate of the batteries. This test determines the battery's ability to hold a charge for a lengthy period.

2.7 Development of batteries

Throughout the progression of our research, we constructed and evaluated fifteen distinct battery prototypes employing a variety of methodologies as in Table 1. After rigorous testing and comprehensive analysis, our investigation culminated in the identification of the most auspicious battery variants, denoted as "Test Battery 13" and "Test Battery 15"

Test Battery	Cell Assembly
1	Electrode : Expanded graphite electrode. Electrolyte : Activated carbon Mixture. Separator : Filter Paper
2	Electrode : Expanded graphite electrode. Electrolyte : Activated carbon Mixture. Separator : KN 95 mask. Current Collector: Copper coated PCB sheet
3	Electrode : Expanded graphite electrode. Electrolyte : Activated carbon Mixture Separator : Filter paper. Current Collector: Copper coated PCB sheet
4	Electrode : Expanded graphite electrode. Electrolyte : Activated carbon Mixture. Separator : Lead Acid battery separator. Current Collector : Copper coated PCB sheet

Table 1: Development of test batteries

5	Electrode : Expanded graphite electrode. Electrolyte : Activated carbon Mixture. Separator : Lead Acid battery separator. Current Collector : Copper coated PCB sheet
6	Electrode : Expanded graphite electrode. Electrolyte : Activated carbon Mixture. Separator : Lead Acid battery separator. Current Collector : Copper coated PCB sheet
7	Electrode : Expanded graphite electrode. Electrolyte : Activated carbon Mixture. Separator : Filter paper. Current Collector: Copper coated PCB sheet
8	Electrode : Expanded graphite electrode. Electrolyte : Activated carbon Mixture. Separator : Lead Acid battery separator. Current Collector : Stainless steel plates
9	Electrode : Expanded graphite electrode. Electrolyte : Activated carbon Mixture. Separator : Lead Acid battery separator. Current Collector: Aluminum plates
10	Electrode : Expanded graphite electrode. Electrolyte : Activated carbon Mixture. Separator : Lead Acid battery separator. Current Collector : Stainless-steel plates
11	Electrode : Activated carbon coated Aluminum sheet. Electrolyte : Phosphoric Acid Separator : Filter Papers
12	Electrode: Sieving mesh with the coated activated carbon Expanded graphite. Electrolyte: Na2SO4 solution. Separator: Filter Paper
13	Electrode : Expanded graphite electrode. Electrolyte : NaCl, NaHCO3, H2SO4, and H3PO4 solutions. Separator : Serviette papers Current Collector : Aluminum plates
14	Electrode : Expanded graphite electrode. Electrolyte : NaCl, NaHCO3, H2SO4, and H3PO4 solutions. Separator : Serviette papers Current Collector : Aluminum plates
15	Electrode : Expanded graphite electrode. Electrolyte : NaCl, NaHCO3, H2SO4, and H3PO4 solutions. Separator : Serviette papers Current Collector : Aluminum plates

2.7.1 Test battery 13

In the 13th test battery configuration, aluminum is employed as the current collector material. To prevent direct contact between the anode and cathode, serviette papers are utilised as separator material.

- Aluminum sheet thickness: 0.5 mm
- Graphite Electrode thickness: 3 mm
- Separator thickness: 1 mm
- Battery size: $5 \operatorname{cm} x 5 \operatorname{cm}$



Figure 4: Test battery 13 graphical illustration

2.7.2 Test battery 15

Building upon the positive outcomes of Test Battery 13, we proceeded with its further development into а six-cell battery configuration. Through meticulous engineering and testing, we successfully assembled six-cell battery the and systematically collected comprehensive data to assess its performance.

Voltage: 3.6 v

Battery Current 24 mA



Figure 5: Representation of one cell of battery 15



Figure 6: Six cell battery illustration.

2.8 Resistance of the graphite electrodes

The resistance of the electrodes was measured by taking measurements from the edges of the electrodes, and subsequently, an average value was calculated.



Figure 7: Points of the electrode where the reading was taken by the multimeter.

2.9 Resistance of the aluminum plates

The resistance of the aluminum plates was assessed by measuring values obtained from the edges of the plates. Subsequently, an average resistance value was computed.



Figure 8: Points of the aluminium plate where the reading was taken by the multimeter.

3 Results

3.1 Resistance of the graphite electrodes and aluminium plates

<i>Table 2: Resistance of the graphite electrodes</i>			
Electrode point	Resistance (Ohm)		
Electrode 1-point X	4.4		
Electrode 1-point Y	4.6		
Electrode 2-point X	4.5		
Electrode 2-point Y	4.3		
Electrode 3-point X	4.7		
Electrode 3-point Y	4.2		
Electrode 4-point X	4.1		
Electrode 4-point Y	4.6		
Electrode 5-point X	4.4		
Electrode 5-point Y	4.3		

The mean resistance values for graphite electrodes are shown in the above table.

4.41

Table 3: Resistance of the Aluminium plates

Average resistance

Aluminum plate point	Resistance (Ω)
Plate 1-point X	5
Plate 1-point Y	5.1
Plate 2-point X	5.2
Plate 2-point Y	4.8
Plate 3-point X	5
Plate 3-point Y	4.9
Plate 4-point X	4.7
Plate 4-point Y	5.2
Plate 5-point X	4.8
Plate 5-point Y	4.6
Plate 6-point X	5.1
Plate 6-point Y	4.7
Plate 7-point X	4.9
Plate 7-point Y	5
Plate 8-point X	4.8
Plate 8-point Y	4.9
Average resistance	4.91875

3.2 Battery capacity

Capacity (in milliampere-hours, mAh) = Current (in milliamperes, mA) x Time (in hours)

Time battery can sustain the 24mA = 36H

Capacity = 24x36

= 864 mAh

3.3 Reactions

The operational principle can be described through the following reactions that are relevant to alkaline electrolytes.

Anodic half reaction Al + $3OH \rightarrow Al (OH)_3$ + $3e^-$ (potential at pH 14: -2.31)

Cathodic half reaction $O_2 + 2H_2O + 4e^- \rightarrow 4OH^-$ (potential at pH 14: +0.40)

Overall reaction $4Al + 3O_2 + 6H_2O \rightarrow 4Al$ (OH)₃ (standard open-circuit voltage: 2.71 V)

3.4 Test battery 15 Self-discharge test

To assess the behavior of the battery over time, a voltage decay test was conducted. The objective of this test was to monitor and quantify the rate at which the battery's voltage decreases over a specified duration.



Figure 9: Self discharge with time

3.5 Test battery 15 Capacity test

To evaluate the capacity of the battery, it was connected to an LED bulb, and a voltage and current testing circuit was employed. This setup allowed for the measurement of both the battery's voltage and the corresponding current flowing through the circuit.



Figure 10: Test battery capacity testing schematic diagram



Figure 11: Voltage varying with time.

5 Discussion

This study presents the characterisation and analysis of a final tested battery with a nominal voltage of 3.6 V and a capacity of 864mAh. Although the battery demonstrates promising performance, it exhibits certain limitations that hinder its usability long-term and recharging capabilities. Specifically, the aluminum plate employed as the current collector suffers from corrosion due to the formation of an Al(OH)₃ layer over time, necessitating periodic replacements. Additionally, the battery is not rechargeable but can be rendered reusable through the substitution of the corroded aluminum plate. Moreover, the electrolyte within the battery depletes over approximately 30 days, necessitating development of the an innovative mechanism to refill the drained electrolyte.

To address these limitations, two potential solutions have been identified: (1) replacing the current aluminum-based electrolyte with Al(Cl)₃, and (2) substituting the aluminum current collector with a copper plate. These solutions have been evaluated to offer several advantages over the current system, specifically in terms of mitigating the corrosion issue and improving the battery's reusability and replenishment of electrolyte. Comprehensive experimentation and comparative analysis have been conducted to assess the feasibility efficacy of proposed and each improvement.

Results indicate that the implementation of Al $(Cl)_3$ as the electrolyte or transitioning to

a copper current collector effectively the corrosion phenomenon, eliminates extending the battery's operational lifespan. Furthermore, the incorporation of a refill mechanism ensures a sustainable supply of electrolyte, allowing for prolonged battery performance without necessitating complete replacements. This study sheds light on the potential advancements that can significantly enhance the reliability and usability of high-capacity batteries, thereby contributing to the sustainable development of energy storage technologies.

5 Conclusion

The characterisation and analysis of a tested battery with a nominal voltage of 3.6V and a capacity of 864 mAh are covered in this paper. Although the battery performs admirably, problems like electrolyte depletion and aluminum current collector corrosion limit the battery's long-term durability. There are two possible fixes suggested: using Al(Cl)₃ as the electrolyte and using copper in place of aluminum for current collector. the According to experimental findings, these strategies successfully reduce corrosion, increase the battery's usable life, and maintain the electrolyte supply. In order to overcome and obstacles advance sustainable alternatives, the report emphasises the significance of ongoing research in energy storage technology.

Furthermore, this study lays the groundwork for enhancing the overall reliability and usability of high-capacity batteries, contributing to the progress towards a cleaner, greener, and more sustainable energy future. With the potential solutions identified and the research paths delineated, we remain optimistic about the bright prospects for overcoming the existing limitations and revolutionising the energy storage landscape.

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References

- I. Hadjipaschalis, A. Poullikkas, and V. Efthimiou, "Overview of current and future energy storage technologies for electric power applications," *Renewable and Sustainable Energy Reviews*, vol. 13, no. 6–7. pp. 1513–1522, Aug. 2009. doi: 10.1016/j.rser.2008.09.028.
- [2] R. Asghar, K. Ullah, Z. Ullah, A. Waseem, N. Ali, and K. Zeb. "Assessment of the Performance and Shortcomings of Common Electric Vehicle Battery Technologies," in 2021 International Conference on Electrical, Communication, and Computer Engineering (ICECCE), IEEE, Jun. 2021. doi: 1-6.pp. 10.1109/ICECCE52056.2021.9514256.
- M. Li, J. Lu, Z. Chen, and K. Amine, "30 Years of Lithium-Ion Batteries," *Advanced Materials*, vol. 30, no. 33, Aug. 2018, doi: 10.1002/adma.201800561.
- [4] C. M. Costa, J. C. Barbosa, R. Gonçalves, H. Castro, F. J. Del Campo, and S. Lanceros-Méndez, "Recycling and environmental issues of lithiumion batteries: Advances, challenges and opportunities," *Energy Storage Mater*, vol. 37, pp. 433–465, May 2021, doi: 10.1016/j.ensm.2021.02.032.
- [5] A. R. Dehghani-Sanij, E. Tharumalingam, M. B. Dusseault, and R. Fraser, "Study of energy storage systems and environmental challenges of batteries," *Renewable and Sustainable Energy Reviews*, vol. 104, pp. 192–208, Apr. 2019, doi: 10.1016/j.rser.2019.01.023.

- [6] Y.-X. Yao, C. Yan, and Q. Zhang, "Emerging interfacial chemistry of graphite anodes in lithium-ion batteries," *Chemical Communications*, vol. 56, no. 93, pp. 14570–14584, 2020, doi: 10.1039/D0CC05084A.
- [7] C. Y. Chan, P. K. Lee, Z. Xu, and D. Y.
 W. Yu, "Designing high-power graphite-based dual-ion batteries," *Electrochim Acta*, vol. 263, pp. 34–39, Feb. 2018, doi: 10.1016/j.electacta.2018.01.036.
- [8] P. Touzain, N. Balasooriya, K. Bandaranayake, and C. Descolas-Gros, "Vein graphite from the bogala and kahatagaha-kolongaha mines, Sri Lanka: A possible origin," *Can Mineral*, vol. 48, no. 6, pp. 1373–1384, Dec. 2010, doi: 10.3749/canmin.48.5.1373.
- [9] L. Fransson and Fyris-tr.), Design and characterisation of new anode materials for lithium-ion batteries. Acta Universitatis Upsaliensis, 2002.
- [10] Y.-J. Cho, I.-J. Park, H.-J. Lee, and J.-G. Kim, "Aluminum anode for aluminum-air battery Part I: Influence of aluminum purity," J Power Sources, vol. 277, pp. 370–378, Mar. 2015, doi: 10.1016/j.jpowsour.2014.12.026.
- [11] N. Soltani, A. Bahrami, L. Giebeler, T. Gemming, and D. Mikhailova, "Progress and challenges in using sustainable carbon anodes in rechargeable metal-ion batteries," *Prog*

Energy Combust Sci, vol. 87, p. 100929, Nov. 2021, doi: 10.1016/j.pecs.2021.100929.

- [12] "399-Development of Sri Lankan Vein Graphite For Lithium-Ion Rechargeable Battery Anodes by Chemical Oxidation".
- [13] P. Murugan, R. D. Nagarajan, B. H. Shetty, M. Govindasamy, and A. K. Sundramoorthy, "Recent trends in the applications of thermally expanded graphite for energy storage and sensors - a review," *Nanoscale Advances*, vol. 3, no. 22. Royal Society of Chemistry, pp. 6294–6309, Nov. 21, 2021. doi: 10.1039/d1na00109d.
- [14] D. Zhang, C. Tan, W. Zhang, W. Pan,
 Q. Wang, and L. Li, "Expanded Graphite-Based Materials for Supercapacitors: A Review," *Molecules*, vol. 27, no. 3. MDPI, Feb. 01, 2022. doi: 10.3390/molecules27030716.
- [15] D. D. L. Chung, "A review of exfoliated graphite," J Mater Sci, vol. 51, no. 1, pp. 554–568, Jan. 2016, doi: 10.1007/s10853-015-9284-6.
- [16] A. M. Haregewoin, A. S. Wotango, and B.-J. Hwang, "Electrolyte additives for lithium ion battery electrodes: progress and perspectives," *Energy Environ. Sci.*, vol. 9, no. 6, pp. 1955– 1988, 2016, doi: 10.1039/C6EE00123H.