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Original Article

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Sustainable Electrochemical Recycling of Spent Zinc–Carbon and Alkaline Batteries to Produce Graphene for High-Performance Supercapacitors

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Abstract: *This paper demonstrates a simple and environmentally friendly approach for recycling spent batteries to produce graphene materials for supercapacitor applications. Using electrochemical exfoliation, graphene was successfully synthesized from the graphite cores of discarded zinc–carbon and alkaline batteries. The synthesized graphene was subsequently employed as an active electrode material for symmetric supercapacitors. Structural characterization via XRD, SEM, EDX, and Raman spectroscopy confirms that the recycled graphene exhibits a well-defined layered structure, high carbon purity (> 80 wt%), large specific surface area, and appropriate surface chemistry. Electrochemical measurements demonstrate that supercapacitors based on recycled graphene electrodes exhibit near-ideal capacitive behavior, rapid charge–discharge response, and excellent cycling stability with over 90% capacitance retention after 1,000 charge–discharge cycles. Compared with conventional battery recycling methods such as mechanical separation, pyrometallurgy, and hydrometallurgy, the proposed electrochemical exfoliation approach offers clear advantages including lower energy consumption, reduced chemical usage, and the production of high-value nanomaterials. This approach not only contributes to mitigating environmental pollution associated with hazardous waste from spent batteries but also enables the production of high-value graphene at significantly lower cost than commercial materials, aligning with circular economy principles and supporting the development of sustainable energy storage systems.*

Keywords: *Graphene, Battery Recycling, Electrochemical Exfoliation, Circular Economy, Sustainable Materials, Supercapacitor.*

INTRODUCTION

The rapid growth of portable electronic devices over recent decades has led to a significant increase in the generation of spent batteries. According to Linden and Reddy [1], billions of batteries are discarded annually, many of which contain heavy metals and hazardous chemical compounds that can cause serious environmental pollution if not properly managed. On a global scale, it is estimated that approximately 15 billion batteries are consumed each year; however, the recycling rate remains extremely low, accounting for only a small fraction of the total waste generated [2].

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In Vietnam, spent batteries are predominantly disposed of through landfilling or incineration together with municipal solid waste. Such disposal practices pose a high risk of releasing heavy metals such as Hg, Cd, and Pb into soil and water systems, while also resulting in the loss of valuable carbon resources contained in the graphite cores of used dry batteries and the missed opportunity for their recovery and reuse. Therefore, the development of recycling strategies that simultaneously mitigate environmental pollution and recover value-added materials has become an urgent necessity.

Concurrently with these environmental concerns, the demand for efficient energy storage devices has increased rapidly due to the expansion of renewable energy systems and modern electronic technologies. Among various energy storage technologies, supercapacitors have attracted considerable attention owing to their high-power density, rapid charge–discharge capability, and long cycle life compared to conventional batteries [3, 4]. The performance of supercapacitors is strongly dependent on the properties of electrode materials, particularly their specific surface area and electrical conductivity.

Graphene, a two-dimensional carbon allotrope with a honeycomb lattice structure, possesses an exceptionally high theoretical specific surface area (~2630 $m^2\ g^{-1}$), excellent electrical conductivity, and outstanding mechanical strength. These characteristics make graphene a highly promising electrode material for supercapacitors [5, 6]. However, commonly used graphene synthesis methods, such as the modified Hummers method, involve strong oxidizing agents and complex chemical processes, raising concerns regarding environmental impact, safety, and production cost [7].

In this context, electrochemical exfoliation of recycled graphite has emerged as a "green" and environmentally friendly approach with strong potential for large-scale production and commercial viability [8]. The utilization of graphite cores from spent batteries as a precursor for graphene synthesis not only contributes to the treatment of hazardous waste but also significantly reduces the cost of producing nanomaterials. Recent studies have demonstrated that graphene derived from recycled carbon sources can still exhibit suitable structural and electrochemical properties for supercapacitor applications [9][10].

Based on these considerations, the present study focuses on the synthesis of graphene from the graphite cores of spent batteries via direct electrochemical exfoliation and the evaluation of its applicability as an electrode material for supercapacitors. This approach aims to integrate environmental protection with the development of sustainable energy storage materials.

EXPERIMENTAL METHODS

Recovery and Pretreatment of Graphite

Spent zinc–carbon and alkaline batteries were collected and manually dismantled. The graphite cores were separated from other battery components, washed repeatedly with deionized water to remove residual electrolytes and chemical impurities, and dried at 80 °C until constant weight was achieved.

Electrochemical Exfoliation of Graphene

Graphene was synthesized via electrochemical exfoliation using a two-electrode system. The recycled graphite rod served as the anode, and a stainless-steel plate served as the cathode. An aqueous ammonium sulfate ($(NH_4)_2SO_4$) solution with a concentration of 0.1 M was employed as the electrolyte due to its chemical stability and environmental compatibility. Based on preliminary optimization experiments, a constant DC voltage of 8 V was applied for 90 min at room temperature. Lower voltages resulted in limited exfoliation efficiency, whereas higher voltages induced excessive oxidation and structural damage. Under the optimized conditions, electrolyte ion intercalation and electrochemical expansion facilitated the effective exfoliation of graphene sheets from the bulk graphite.

Electrode Fabrication and Material Characterization

After exfoliation, the graphene product was filtered, thoroughly washed with deionized water to remove residual electrolyte, and dried to obtain graphene powder. The material was then processed into electrode films and assembled into supercapacitor devices for electrochemical evaluation.

The crystalline structure of graphene was characterized by X-ray diffraction (XRD) over a 2θ range of 10°–80°, showing a characteristic interlayer spacing of approximately 0.34 nm. The morphology and structural features of the graphene were examined using scanning electron microscopy (SEM) and transmission electron microscopy (TEM), while its electrical and electrochemical properties were measured using cyclic voltammetry (CV) and electrochemical impedance spectroscopy (EIS) to assess its suitability for supercapacitor applications.

RESULTS AND DISCUSSION

Electrode Fabrication and Material Characterization

The results demonstrate that electrochemical exfoliation effectively converts bulk graphite recovered from spent batteries into graphene sheets with high purity. The synthesized graphene preserves the characteristic sp^2-bonded honeycomb lattice, ensuring good electrical conductivity and a large electrode–electrolyte interfacial area, which is crucial for supercapacitor applications [5].

The XRD pattern (Figure 1) shows a distinct diffraction peak at 2θ ≈ 26.3°, corresponding to the (002) plane of graphitic carbon with an interlayer spacing of approximately 0.34 nm.

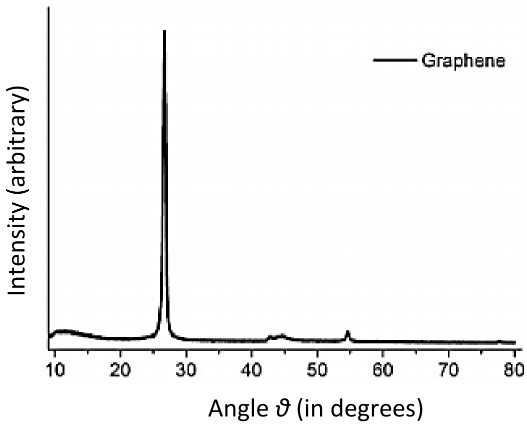
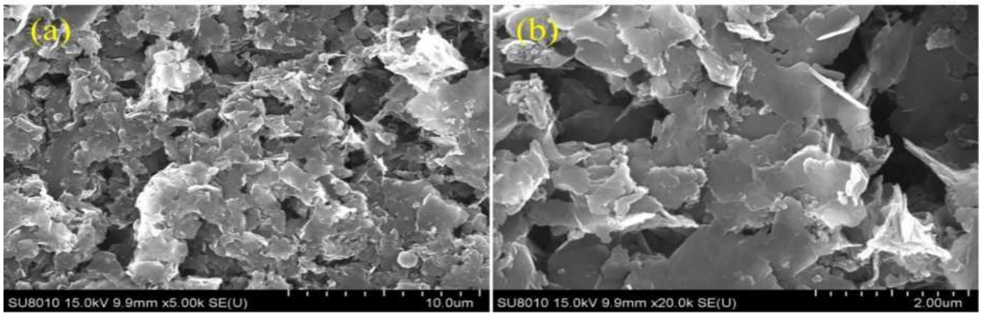


Figure 1: X-ray diffraction (XRD) results of graphene material.

SEM images (Figures 2a and 2b) reveal thin, loosely stacked graphene sheets with slightly curled edges, a typical morphology of electrochemically exfoliated graphene. At higher magnification, the sheets exhibit smooth surfaces with lateral dimensions exceeding 2 µm, indicating efficient exfoliation and good control of electrochemical parameters. These features suggest the predominance of few-layer graphene, consistent with the XRD analysis.

EDX analysis (Figure 3) indicates that carbon is the major constituent (> 80 wt%), while oxygen (10–15 wt%) originates from oxygen-containing functional groups that enhance electrolyte wettability and ion adsorption capacity. A small nitrogen content, likely introduced during electrochemical processing, may act as a dopant and contribute to enhanced electrical conductivity and electrochemical activity.



(a) (b)

Figure 2: SEM images of graphene material at different magnifications: (a) 5,000× and (b) 20,000×.

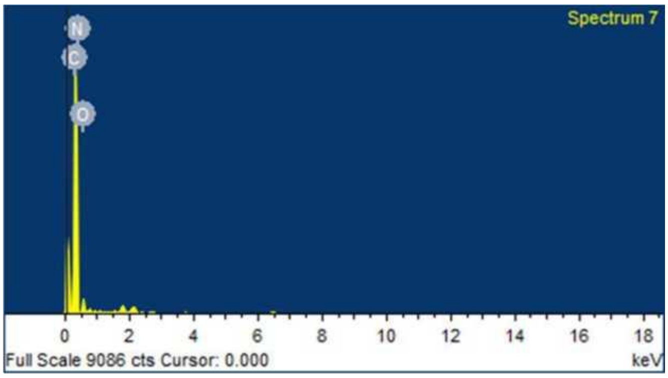


Figure 3: Energy-dispersive X-ray (EDX) spectrum results of graphene material.

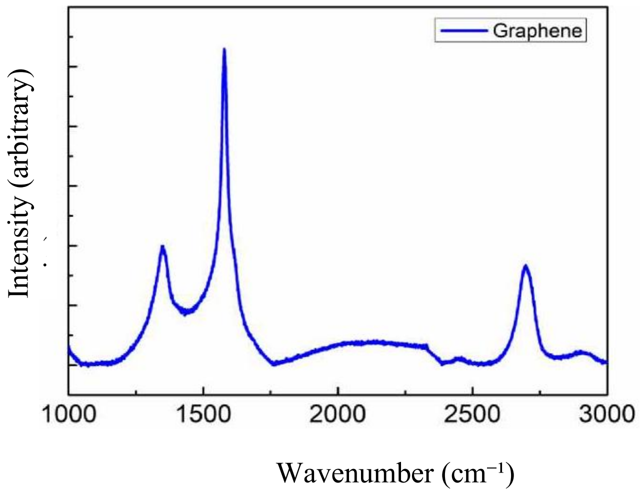


Figure 4: Raman spectrum results of graphene material synthesized from waste battery cores.

Raman spectra (Figure 4) display the characteristic D (~1350 cm^{-1}), G (~1580 cm^{-1}), and 2D (~2700 cm^{-1}) bands of graphene. The ID/IG ratio of approximately 0.4 indicates a moderate defect density, providing a favorable balance between structural integrity and active sites for electrochemical reactions, in agreement with previous studies [11]. Overall, the combined

XRD, SEM, EDX, and Raman results confirm that graphene derived from spent batteries possesses a well-defined layered structure, high carbon content, and suitable surface chemistry, meeting the key requirements for supercapacitor electrode materials [12].

Fabrication of Supercapacitor Electrodes

Supercapacitor electrodes were fabricated using graphite paper (1 × 2 cm, thickness 0.8 mm) as the current collector. The active layer consisted of synthesized graphene, activated carbon, and PVDF binder in a mass ratio of 80:10:10, using N-methyl-2-pyrrolidone (NMP) as the solvent. The homogeneous slurry was coated onto the graphite paper over an active area of 1 × 1 cm and dried at 40 °C to remove solvent and improve adhesion prior to electrochemical testing.

Electrochemical Performance

The electrochemical performance of symmetric supercapacitors assembled with recycled graphene electrodes was evaluated using cyclic voltammetry (CV) and galvanostatic charge–discharge (GCD) measurements. The CV curves (Figure 5a) exhibit a near-rectangular shape over a wide range of scan rates, indicating dominant electric double-layer capacitance with a contribution from pseudocapacitive surface reactions. This behavior reflects good rate capability and electrochemical stability.

The galvanostatic charge–discharge (GCD) curves recorded at current densities of 0.25 and 0.5 A g^{-1} (Figure 5b) display nearly symmetric triangular shapes, confirming ideal capacitive behavior and high Coulombic efficiency. Cycling tests reveal that the specific capacitance remains stable and comparable to that of commercial carbon materials, with more than 90% capacitance retention after 1,000 charge–discharge cycles. These results demonstrate the fast response and excellent durability of supercapacitors based on recycled graphene electrodes.

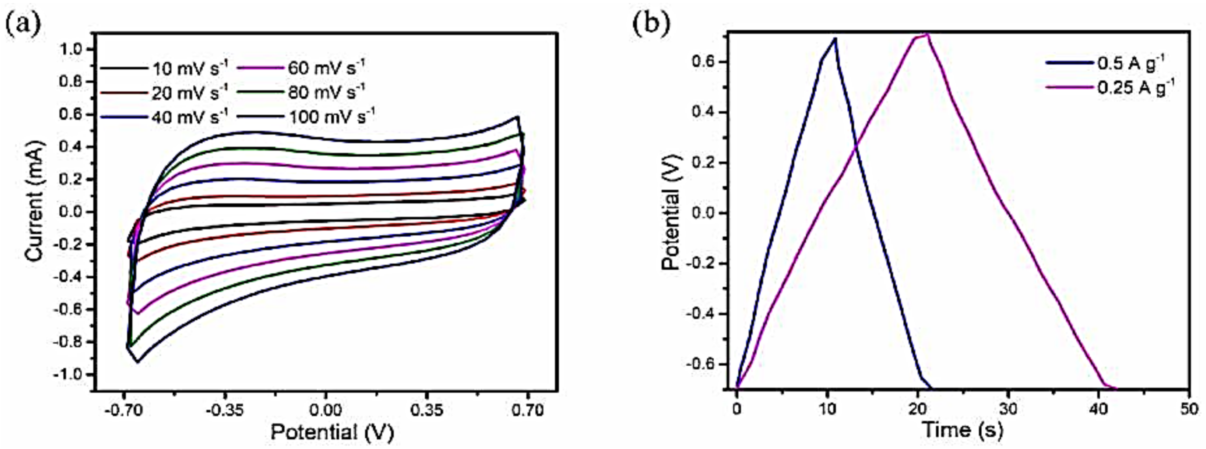


Figure 5: (a) Cyclic voltammetry curves at different scan rates; (b) charge–discharge curves at different current densities.

Discussion

Compared with conventional battery recycling methods such as mechanical separation, pyrometallurgy, and hydrometallurgy, electrochemical exfoliation offers clear advantages, including lower energy consumption, reduced chemical usage, and the production of high-value graphene materials [13]. The ability to control graphene thickness and quality through applied voltage and exfoliation time enables the preparation of few-layer graphene with tailored electrochemical properties.

Beyond supercapacitor applications, graphene recovered from spent batteries exhibits potential for use in lithium-ion battery electrodes, sensors, conductive composites, and environmental remediation applications [14]. The Life-cycle assessment studies further indicate that supercapacitors employing graphene-based electrodes can significantly reduce environmental impact compared with conventional systems [15]. Importantly, the use of recycled graphene substantially lowers electrode material costs relative to commercial graphene while maintaining the required electrochemical performance. This cost–performance advantage aligns well with circular economy principles based on a "collection–recycling–manufacturing–application" framework and demonstrates the feasibility of small-scale, localized battery recycling systems that transform hazardous waste into valuable resources for advanced technological applications.

CONCLUSION

This work demonstrates a simple and environmentally friendly approach for recycling spent batteries into graphene suitable for supercapacitor applications. Graphene was successfully synthesized from discarded battery graphite via electrochemical exfoliation, yielding few-layer graphene with high carbon purity and appropriate surface chemistry. Structural characterization confirms the preservation of the graphitic layered structure, while electrochemical measurements reveal near-ideal capacitive behavior, rapid charge–discharge response, and excellent cycling stability with over 90% capacitance retention after 1,000 cycles. Compared with conventional recycling methods, the proposed approach reduces chemical and energy consumption while producing high-value graphene at significantly lower cost than commercial materials. These results highlight the potential of converting battery waste into functional electrode materials and support the development of sustainable energy storage systems within a circular economy framework.

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