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From waste graphite to high-value carbon materials: direct recycling and nanotechnology-enabled upcycling of lithium-ion battery anodes

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Abstract: The direct recycling of graphite, the anode material in lithium-ion batteries, is comprehensively examined in this study. The research integrates mineral processing techniques with a nanostructural regeneration approach. Graphite is a vital component of modern battery technologies due to its high electrical conductivity, chemical stability, and capacity for lithium intercalation. However, traditional pyrometallurgical and hydrometallurgical recycling methods primarily focus on recovering valuable metals, while degrading the graphite structure, which subsequently requires energy-intensive synthesis processes for reuse. The direct recycling approach aims to restore the performance of electrode materials by preserving their crystal structure and morphology. While graphite could recover from black mass with over 90% efficiency via flotation following mechanical pre-treatment, preserving its structural integrity, the process remains inherently complex due to surface impurities, heterogeneous phase composition, limitations in particle liberation and selectivity, and the requirement for precise reagent optimization. Furthermore, the study evaluates relithiation methods for cathodes and purification techniques for graphite anodes, including thermal, chemical, electrochemical, and hybrid approaches. The findings indicate that direct recycling significantly reduces energy consumption and lowers global warming potential by 70–96%, while generating high economic added value. In conclusion, the direct recycling approach offers a strong alternative for sustainable battery technologies from environmental, economic and technological perspectives.

Keywords: direct recycling, graphite, battery technology, nanostructural upcycling, spent lithium-ion batteries

1. Introduction

Graphite is recognized as a strategic resource supporting 21st-century high technologies due to its unique physical and chemical properties. Its combination of metallic properties (such as electrical and thermal conductivity) and non-metallic properties (including high thermal resistance, chemical inertness, and lubricity) makes it indispensable to modern industry. Classified as a "Critical Raw Material" by the European Union, graphite is a key component in essential sectors such as lithium-ion batteries, nuclear reactors, aerospace, and steel production (Kaya et al., 2009; Tian et al., 2024). Fig. 1 shows various graphite applications in the energy sector.

Graphite is a layered allotrope of carbon with a hexagonal crystal structure formed by sp^2 hybridisation. These layers are bonded internally by strong covalent bonds and stacked together by weak van der Waals forces. This crystal architecture provides graphite with high electrical conductivity, chemical stability, and excellent intercalation capacity. Due to these properties, graphite is the most widely used anode material, particularly in lithium-ion batteries (LIBs). Consequently, the recovery and regeneration of graphite from spent lithium-ion batteries have attracted significant attention, prompting the development of various recycling strategies (Han et al., 2024). The rapid growth of electric vehicles, portable electronics, and large-scale energy storage systems has led to an unprecedented increase in the

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production and consumption of lithium-ion batteries worldwide. Consequently, the volume of spent batteries reaching end-of-life is expected to rise substantially in the coming decades. Although recycling efforts have traditionally focused on recovering high-value metals such as lithium, cobalt, and nickel, graphite represents a significant fraction of battery mass and remains a strategically important material due to its widespread use and energy-intensive production. Therefore, the efficient recovery and regeneration of graphite have become increasingly important from both economic and environmental perspectives, driving the development of advanced recycling technologies for spent lithium-ion batteries (Kayakool et al., 2021).

Fig. 1. Schematic illustration of graphite applications in energy storage systems

Spent battery recycling strategies are generally classified into three main categories: pyrometallurgical, hydrometallurgical, and direct recycling approaches (Bai et al., 2020; Yu, Bai, Polzin, et al., 2024). These methods employ various physical, chemical, and electrochemical techniques to recover valuable components from complex electrode materials. For example, commonly reported techniques in lithium-ion battery recycling include acid leaching (Nitta et al., 2015; Yu et al., 2023), direct delamination (Guo et al., 2023), solvent extraction (Kang et al., 2010), hydrothermal treatment (Gao et al., 2023), membrane separation (Wagh et al., 2023), and electrochemical processes (Kim et al., 2021). Pyrometallurgical and hydrometallurgical methods primarily recover valuable elements through thermal or chemical decomposition. In contrast, direct recycling aims to preserve and restore the original structure and functionality of electrode materials without breaking them down into their elemental constituents. This approach offers several advantages, including lower energy consumption, reduced chemical usage, and preservation of the microstructure, physicochemical properties, and surface chemistry of active materials. As a result, direct recycling has emerged as a promising and sustainable strategy, particularly for structurally sensitive materials such as graphite, whose electrochemical performance strongly depends on crystallographic integrity, defect structure, and surface properties (Yu, Bai, & Belharouak, 2024).

Regardless of the recycling strategy, recovering valuable materials from spent lithium-ion batteries requires a sequence of preliminary operations. These initial steps are essential to ensure operational safety, facilitate material separation, and improve the efficiency of downstream recycling processes. Therefore, pre-treatment is generally considered a crucial stage in battery recycling and is commonly applied before pyrometallurgical, hydrometallurgical, or direct recycling routes. Pre-treatment involves discharging the battery to eliminate residual charge, followed by removing external components such as frames, cables, and electronic circuits. Subsequently, individual materials, including plastics, metals, electrolytes, and electrodes, are separated to obtain the black mass containing the active electrode materials (Fig. 2) (Barbosa de Mattos et al., 2025).

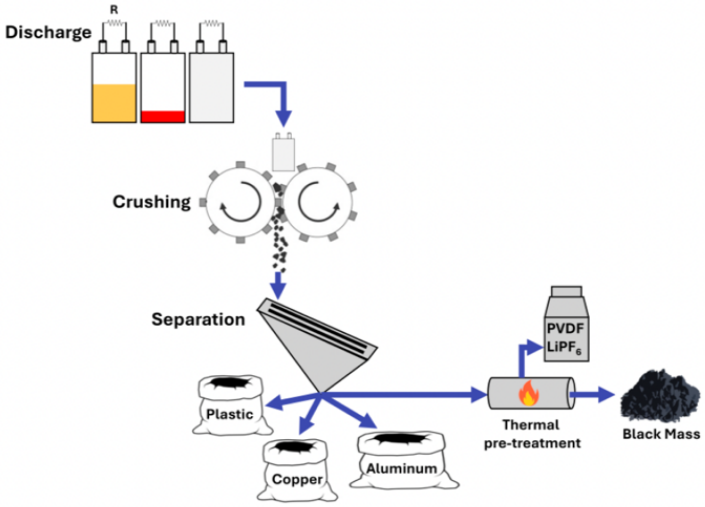

Fig. 2. Illustration of mechanical and separation processes used to recover black mass (Barbosa de Mattos et al., 2025)

Fig. 3 presents a flowchart illustrating the direct recycling process. From a mineral processing perspective, graphite recovery from spent batteries relies primarily on physical separation techniques. Graphite is naturally hydrophobic, enabling efficient separation by froth flotation. During flotation, graphite selectively attaches to air bubbles and separates from hydrophilic cathode materials, such as lithium metal oxides. Optimized flotation conditions have achieved recovery efficiencies exceeding 90%. Flotation is a key mineral processing technique that enables selective separation of graphite from cathode materials after pre-treatment, allowing recovery without structural degradation and facilitating direct recycling (Bhar et al., 2023).

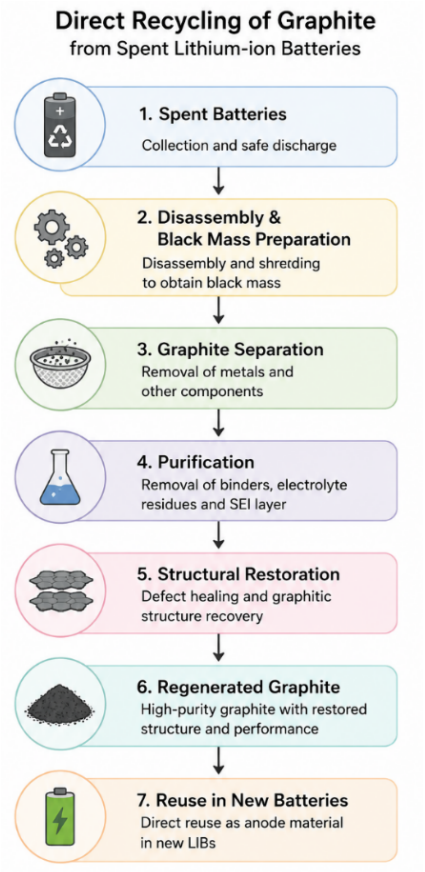

Fig. 3. Flowsheet of the direct recycling process of graphite from spent Lithium-Ion Batteries

2. Types of graphite

Graphite can be broadly divided into two principal categories (natural graphite and synthetic graphite) based on its origin and formation mechanism. Natural graphite forms through geological processes over geological timescales, whereas synthetic graphite is produced by controlled graphitization of carbon-rich precursor materials at elevated temperatures, typically between 2,300 and 3,000 °C. Conventional synthetic graphite is primarily derived from petroleum coke and coal tar pitch, which enable precise control over purity, crystallinity, and particle morphology. In addition to fossil-based precursors, attention has increasingly shifted to sustainable sources such as biomass-derived carbon, including lignin, cellulose, and agricultural waste. These renewable precursors offer environmentally friendly alternatives for producing synthetic graphite with tunable microstructure, defect density, and nanoscale properties, enhanced ion transport pathways, and potential quantum confinement effects at reduced crystallite sizes.

Natural graphite is primarily classified into three main types based on its geological formation and physical structure: microcrystalline (amorphous), vein (lump), and crystalline flake graphite (Simandl et al., 2015). Fig. 4 shows the morphology and size characteristics of different natural graphite types.

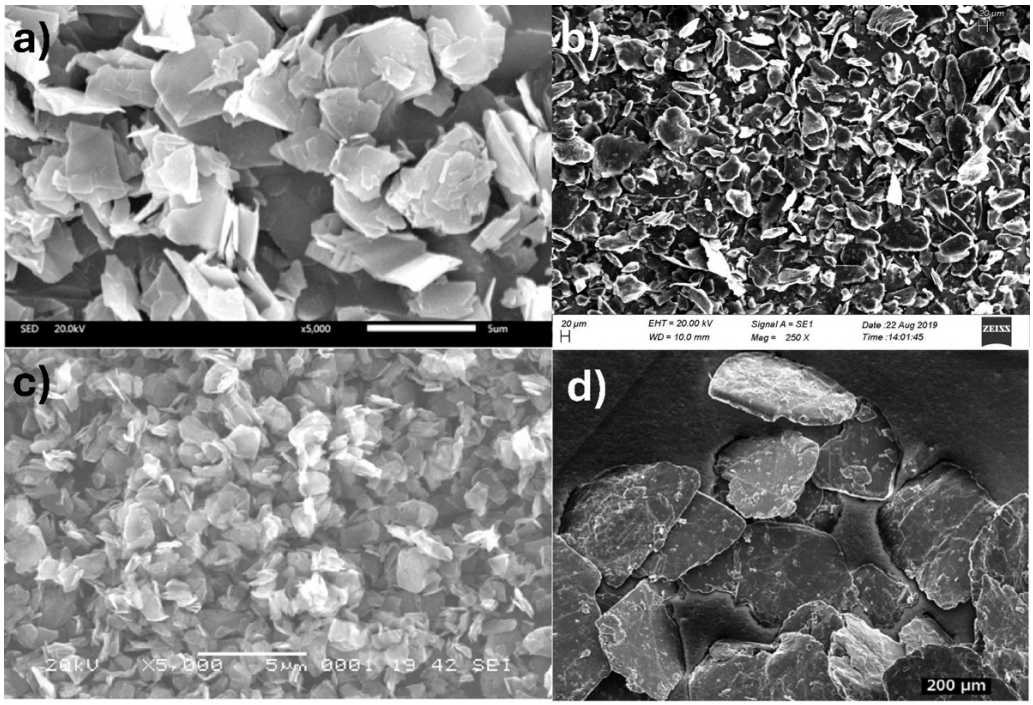

Fig. 4. SEM images illustrating the size and morphology of different graphite types: (a) amorphous graphite (Wang et al., 2017), (b) vein graphite (Somaweera et al., 2022), and (c-d) flake graphite (Lai et al., 2012; Meyer-Plath et al., 2015)

2.1. Flake graphite

Flake graphite occurs as flat, sheet-like particles within metamorphic rocks such as schist, marble, and gneiss. It exhibits a layered, anisotropic structure, with high electrical and thermal conductivity parallel to the basal planes, while conductivity perpendicular to these planes is significantly lower. Flake graphite demonstrates high reversible capacity and high initial Coulombic efficiency (ICE), making it highly suitable for electrochemical energy storage applications. Its carbon purity typically ranges from 85% to 97%. However, higher purities exceeding 99% can be achieved through beneficiation and purification processes, which makes it the most critical graphite type for advanced technological applications. Through mechanical processing, flake graphite can be transformed into spherical graphite, which is particularly suitable for use as an anode material in lithium-ion batteries. Beyond battery applications, flake graphite is widely used in refractory materials and lubricants because of its excellent thermal stability and lubricating properties (Dehaine et al., 2020; Kalyoncu et al., 2014; Simandl et al., 2015).

2.2. Vein or lump graphite

Vein graphite is found in rock fractures within host rocks, typically forming vein-like or pipe-like structures. It is generally considered to be of hydrothermal origin. This graphite type has a massive structure and is among the purest naturally occurring forms of graphite, with carbon contents ranging from 90% to 99%. The world's most significant deposits are located in Sri Lanka. Because of its high purity and excellent electrical conductivity, vein graphite is commonly used in specialised carbon products and electrical components such as motor brushes. Although it was historically used extensively in refractory applications, flake graphite has largely replaced it in this sector (Dehaine et al., 2020; Kalyoncu et al., 2014).

2.3. Microcrystalline (amorphous) graphite

Microcrystalline graphite forms through the metamorphism of coal deposits. The term 'amorphous' (meaning 'shapeless') is a commercial designation, as this material consists of very small crystallites with low crystallinity rather than a truly amorphous structure. It typically contains a higher percentage of ash and mineral impurities, which can be difficult to remove using conventional beneficiation processes. It exhibits relatively isotropic properties at the macroscopic scale and demonstrates good cycle stability due to its lower volumetric expansion during electrochemical cycling. Microcrystalline graphite is commonly used as a carbon additive in steel production, foundry coatings, lubricants, and pencil manufacturing (Beyssac and Rumble, 2014; Kalyoncu et al., 2014; Simandl et al., 2015).

2.4. Synthetic graphite

In addition to natural graphite, synthetic graphite is produced through the high-temperature graphitization of carbon-rich precursor materials such as petroleum coke and coal tar pitch. This process typically occurs at temperatures between 2,300 and 3,000 °C, resulting in highly ordered graphite structures. Synthetic graphite generally exhibits higher purity, improved structural uniformity, and longer cycle life compared to natural graphite. However, its production is energy-intensive and associated with significantly higher costs (Dehaine et al., 2020).

2.5. Expanded graphite and graphene

Graphite also serves as a precursor material for advanced carbon materials such as expanded graphite and graphene. Expanded graphite is produced by chemically treating flake graphite followed by rapid thermal expansion, which causes separation and expansion of the layered structure. It is widely used in sealing products, thermal management systems, and conductive foils. Graphene, a single atomic layer of graphite, possesses exceptional electrical, thermal, and mechanical properties. Often referred to as a 'miracle material', graphene has significant potential in advanced applications, including energy storage, sensors, electronics, biomedical devices, and additive manufacturing technologies.

3. Importance of direct recycling of graphite for battery technologies

Graphite plays a critical role as the primary anode material in modern battery technologies, particularly in lithium-ion batteries (LIBs). Its widespread use is primarily attributed to its low lithium intercalation/extraction potential and excellent electrical conductivity. During battery operation, lithium ions intercalate between graphite layers to form LiC_6, which provides a theoretical specific capacity of 372 mAh/g and a volumetric capacity of approximately 840 mAh/cm^3 (Dusastre, 2001). Graphite maintains its structural stability during repeated charge and discharge cycles, enabling efficient Lithium-ion diffusion and long cycle life. Natural flake graphite is commonly processed into spherical graphite to improve packing density, electrode uniformity, and electrochemical performance.

In terms of market share, synthetic graphite accounts for 58% of battery-grade graphite production, while natural graphite represents approximately 39% (Zhao et al., 2022a). Natural graphite is preferred because it has a lower production cost, better processability, and a smaller environmental footprint than synthetic graphite. While flake graphite offers high capacity, microcrystalline graphite exhibits improved rate capability under certain conditions. Synthetic graphite, offering higher purity and longer cycle life, is associated with higher energy consumption and production costs (Wu et al., 2002; Yazici et

al., 2005). To be used as an anode material in lithium-ion batteries, natural graphite must be converted into spherical graphite through a series of physical and chemical processes. This involves transforming crystalline flake graphite into high-purity, spherical particles suitable for electrochemical applications (Huang et al., 2001). Crystalline flake graphite concentrate is typically used as the primary raw material for producing spherical graphite, with an initial carbon content of approximately 95% (Bai et al., 2013; Huang et al., 2001). The flakes are mechanically reduced in size and reshaped into rounded particles resembling 'potatoes', rather than retaining their original flake morphology. The flexible layered structure of graphite enables these particles to be transformed into spherical shapes. For efficient battery performance, graphite must be very pure. Therefore, the mechanically shaped graphite undergoes chemical purification processes to increase its carbon content to approximately 99.9%. In the final stage, surface coating treatments are applied to enhance the electrochemical performance and cycle life of the spheroidised and purified graphite. Spherical graphite is a critical material in lithium-ion technology due to its superior performance as an anode material, and its demand rapidly increasing due to the growth of electric vehicle market (Luo et al., 2022; Tian et al., 2024; Zhao et al., 2022a). The rapidly growing demand for battery-grade graphite, driven by the expansion of the electric vehicle and energy storage sectors, has raised concerns regarding raw material supply, production costs, and environmental sustainability. Although natural and synthetic graphite are essential components of modern lithium-ion batteries, extracting and processing them requires considerable energy and resources. Consequently, recovering and regenerating graphite from spent lithium-ion batteries has emerged as an attractive strategy to reduce dependence on primary resources, mitigate environmental impacts, and support the development of a circular battery economy.

Lithium-ion battery recycling has become increasingly important, and methods are generally classified into three main categories: pyrometallurgical, hydrometallurgical, and direct recycling (Xu et al., 2023). Pyrometallurgical recycling is a well-established approach with high processing capacity that involves melting batteries in furnaces at approximately 1400 °C (Abdollahifar et al., 2023; Chernyaev et al., 2024). In hydrometallurgical recycling, batteries are processed to obtain 'black mass', and strong acids such as sulphuric acid dissolve valuable metals into a liquid phase (Abdollahifar et al., 2023; Tokoro et al., 2025). Metal salts (such as lithium carbonate, nickel sulfate) are then recovered through processes including filtration, precipitation, and solvent extraction. Although hydrometallurgical methods can recover lithium more effectively than pyrometallurgical methods, they have disadvantages, including high chemical consumption, complex wastewater treatment requirements, and the need for material resynthesis (Xu et al., 2023). Direct recycling of lithium-ion battery components, including natural and synthetic graphite, is an economical and environmentally friendly method that aims to restore spent electrode materials to their original performance without completely breaking down their structure, thereby preserving their morphology and crystal structure (He et al., 2025). This approach enables reuse of spent battery materials in new battery production by preserving structural integrity and renewing performance through physical and/or chemical treatments, rather than decomposing them into elemental forms. These methods generally involve recovering lithium (relithiation) for cathode materials and purifying/improving anode (graphite) materials (Barre et al., 2025). Advantages and disadvantages of these methods are shown in Fig. 5.

For lithium recovery, the primary cause of capacity loss in cathode materials is lithium loss during charge-discharge cycles. Various methods have therefore been developed to restore the performance of used cathodes, including solid-state sintering, hydrothermal/solution-based re-lithiation, eutectic molten salt, ionic liquid, and electrochemical re-lithiation. These methods primarily aim to restore the cathode material's electrochemical performance by reincorporating lithium ions into the crystalline structure (Xu et al., 2023).

3.1. Anode (graphite) recycling methods

Graphite recovery primarily focuses on removing binders and the solid electrolyte interphase (SEI) layer. Washing processes involve treating graphite powders with various solvents such as dimethyl carbonate (DMC), N-methyl-2-pyrrolidone (NMP), or water to remove binders (such as PVDF or CMC/SBR) and electrolyte residues. Thermal treatment and pyrolysis involve heating materials in an inert atmosphere (usually argon) at temperatures ranging from 600°C to 1000°C, or higher (sometimes

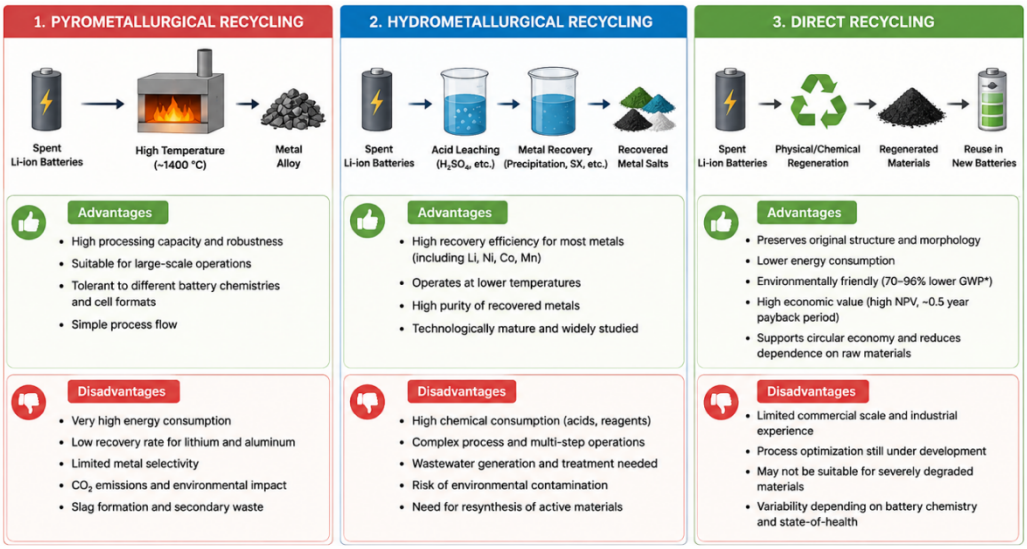

Fig. 5. Comparison of the advantages and disadvantages of pyrometallurgical, hydrometallurgical, and direct recycling methods

up to 2900°C) to vaporize organic binders and restore graphite crystallinity (Abdollahifar et al., 2023; Xu et al., 2023; Zheng et al., 2024). Rapid thermal shock treatment removes impurities and forms an amorphous carbon layer on the surface to improve performance by heating the graphite to very high temperatures (1000–2500 °C) within seconds, then rapidly cooling it. Hybrid methods combining chemical and thermal treatments typically involve removing metal impurities with a mild acid leach (usually a mixture of H_2SO_4 and H_2O_2), followed by thermal restoration. Electrochemical methods involve separating graphite from the copper current collector foil while simultaneously recovering lithium in solution (Abdollahifar et al., 2023; Martin et al., 2026; Muguruza-Sánchez et al., 2025a).

3.2. Advanced regeneration and upcycling techniques

Electrical Pulsed Discharge (EPD) is a physical separation method that uses high-voltage pulses to detach active materials (graphite or cathode materials) from current collector foils (Cu or Al) without heat or chemicals. Surface coating and upcycling techniques involve modifying spent graphite with functional coatings such as calcium silicate (C-S-H) to improve electrochemical performance, particularly fast-charging capabilities. Boron doping treats graphite with boron-containing compounds to enhance electrochemical capacity and conductivity. The indirect recycling approach combines conventional chemical recovery processes with remanufacturing techniques. In this method, battery components are decomposed into elemental or precursor forms, which are then used directly to produce new cathode active materials (CAMs). This approach transforms traditional 'recycling' into a closed-loop material regeneration process (Barbosa de Mattos et al., 2025).

Direct recycling of lithium-ion battery components is an economically and environmentally advantageous approach that aims to directly regenerate spent electrode materials while preserving their crystal structure and morphology, rather than separating them into their elemental components. This process reduces global warming potential by 70–96% compared to conventional recycling methods and significantly lowers energy consumption by preserving the embodied energy of the original materials. In addition, this approach is highly economically feasible, with a high Net Present Value (NPV) and a short payback period of about 0.5 years, because it produces high-value electrode materials without complete re-synthesis. Furthermore, it reduces dependence on external raw material sources by enabling the local recovery of critical raw materials such as graphite and lithium, thereby supporting the development of a closed-loop circular economy for battery technologies.

4. Comparison of graphite recycling strategies based on life cycle assessment

Table 1 compares the three main graphite recycling methods in terms of energy consumption, production costs, global warming potential (GWP), and recovery efficiency. While all three approaches

recover graphite from spent lithium-ion batteries, they differ significantly in environmental impact, economic feasibility, and the quality of the recovered graphite. Hydrometallurgical recycling generally has relatively low energy consumption, ranging from approximately 0.4 to 2.5 kWh per kg of recovered graphite, due to its operation at moderate temperatures. Despite this low energy demand, however, it is the most expensive of the three recycling methods in terms of production costs. This is primarily due to the extensive use of strong acids, oxidising agents, neutralising chemicals, multiple purification steps and wastewater treatment requirements. Furthermore, the generation of secondary liquid waste streams increases operational complexity and processing costs. Nevertheless, hydrometallurgical processes typically achieve high recovery efficiencies (>85%) together with relatively low GWP values (2.5–3 kg CO_2 – eq/kg), making them environmentally attractive when efficient chemical management and wastewater treatment strategies are employed (Premathilake et al., 2025; Rey et al., 2021).

By contrast, pyrometallurgical recycling involves high-temperature thermal treatment, typically at temperatures of 1000–1400 °C or higher. Consequently, its energy demand is substantially higher than that of hydrometallurgical processes and varies with furnace design, process configuration, and energy source. Similarly, the wide range of reported global warming potential (GWP) values (0.5–7 kg CO_2 – eq/kg) primarily reflects variations in fuel consumption, electricity generation, and the system boundaries adopted in life cycle assessment studies. While pyrometallurgical recycling generally incurs lower production costs than hydrometallurgical recycling due to its relatively simple process flow and well-established industrial infrastructure, exposure to high temperatures often damages the original graphite crystal structure. Consequently, the recovered graphite often requires additional purification or graphitisation treatments before it can be reused as an anode material, thereby reducing its overall material value (Premathilake et al., 2025; Rey et al., 2021).

Direct recycling is one of the most promising approaches among the available recycling technologies because it preserves the original crystal structure and morphology of graphite instead of decomposing it into its elemental constituents(Muguruza-Sánchez et al., 2025b). Unlike conventional recycling technologies, direct recycling focuses on removing surface contaminants and the SEI, repairing structural defects, and restoring graphitic ordering to recover electrochemical performance. Recent techno-economic analyses have demonstrated that direct recycling can achieve production costs of around 18 USD per kg while maintaining recovery efficiencies of over 85% and a global warming potential (GWP) of around 4 kg CO_2 – eq/kg. While its carbon footprint is not necessarily lower than that of hydrometallurgical recycling, direct recycling eliminates the need for complete material re-synthesis. This minimises material losses, reduces resource consumption and preserves the intrinsic value of graphite (Gkousis et al., 2026; Lefherz et al., 2025).

A key advantage of direct recycling is that recovered graphite can be reused as an anode material after appropriate restoration treatments. Numerous studies have demonstrated that regenerated graphite can recover electrochemical capacities comparable to commercial graphite, while maintaining excellent cycling stability. Furthermore, nanostructural restoration strategies, including defect healing, graphitic layer reconstruction, surface coating, heteroatom doping, and controlled carbon deposition, further improve electrical conductivity, lithium-ion diffusion kinetics, and electrode interfacial stability. These nanoscale modifications restore the original electrochemical performance of graphite and may enhance its long-term cycling durability beyond that of conventionally regenerated materials (Shen et al., 2023).

Beyond lithium-ion batteries, recycled graphite has shown strong potential for a variety of valuable applications. The structural defects and oxygen-containing functional groups that are generated during the regeneration process provide an abundance of active sites for adsorption and catalytic reactions. Consequently, it has been successfully utilised as an adsorbent for heavy metal removal, a catalyst support for advanced oxidation processes, a supercapacitor electrode material, and a precursor for graphene oxide synthesis. These value-added applications significantly enhance the economic viability of graphite recycling, transforming spent graphite from battery waste into a versatile carbon resource for advanced energy and environmental technologies. Overall, this comparison shows that no single recycling technology is superior in all respects. Hydrometallurgical recycling offers relatively low greenhouse gas emissions but suffers from high chemical consumption and elevated operating costs. Pyrometallurgical recycling is robust and comparatively inexpensive to operate; however, it requires

substantial energy input and often compromises the structural integrity of graphite. In contrast, direct recycling offers a balanced approach, combining environmental sustainability, economic feasibility, and material preservation. By maintaining the original crystal structure and restoring graphite through nanostructural engineering, direct recycling maximises the value of recovered graphite, supporting the development of a circular economy for lithium-ion batteries. As nanotechnology-based restoration strategies evolve, direct recycling is expected to play an increasingly important role in the sustainable manufacturing of batteries and the efficient utilisation of critical carbon resources.

Table 1. Comparison of recycling methods in terms of energy, cost, and environmental performance

Method	*EC (kWh/kg graphite)	Cost (USD/kg *G)	*GWP (kg CO_2 eq/kg *RG)	Recovery (%)	Ref
Hydrometallurgical	0.398-2.5	420	2.5-3	>85	(Kayakool et al., 2021; Premathilake et al., 2025)
Pyrometallurgical	1-10	159	0.5-7	>75	(Premathilake et al., 2025; Rey et al., 2021)
Direct Recycling		18	4	>85	(Gkousis et al., 2026)

5. Nanostructural restoration and reutilization of recycled graphite

The primary goal of recycling graphite from used lithium-ion batteries is not only to physically recover the material but also to restore the structural and electrochemical properties it has lost during use. During battery cycling, SEI residues form on the graphite surface, the layered structure becomes partially disrupted, and some of the active regions lose their functionality (Adenusi et al., 2023). For this reason, recycling strategies developed in recent years focus not only on cleaning the graphite but also on its structural regeneration. The literature often defines this approach as "restoration" or "regeneration" and aims to restore graphite's crystal structure, surface properties, and electrochemical performance (Ji et al., 2024). Direct recycling methods offer significant sustainability advantages because they enable graphite to be reused while preserving its existing structure. Controlling structural changes at the nanoscale plays a critical role in enhancing the reusability of graphite. By eliminating defects formed during use, improving the interlayer arrangement, and reorganizing surface functional groups, graphite's electrical conductivity and lithium storage performance can be significantly enhanced (Zhao et al., 2022b). For this reason, many current studies focus on the reuse of recycled graphite as battery anodes (Moradi and Botte, 2015). Research has shown that waste graphite, when subjected to appropriate regeneration processes, can exhibit capacity values close to those of new graphite and provide long-term cycle stability. This makes it possible to repurpose existing material instead of producing new graphite, which is energy-intensive and costly. This approach both reduces the strain on natural graphite resources and supports the development of a circular economy approach in battery production (Xu et al., 2022). Moreover, the applications of recycled graphite extend beyond the production of new batteries. The functional groups, defects, and active sites that form on its surface during regeneration processes can also offer advantages for a variety of other applications. Recent studies have shown that waste graphite can be used as an adsorbent, a catalyst support, a supercapacitor electrode, and an environmental remediation material. In advanced oxidation processes and Fenton-like reactions (Peng et al., 2025), recycled graphite has shown effective performance in degrading organic pollutants due to active sites on its surface (Deng et al., 2023). Similarly, recycled graphite has been reported to demonstrate successful performance in heavy metal adsorption and wastewater treatment applications (Zhao et al., 2017). These studies demonstrate that graphite is a valuable carbon source that can be utilised across various industries, not merely a recovered battery component.

Peng et al. (2025) aimed to recycle waste lithium-ion battery graphite with different degradation characteristics through a high-value recycling approach. The graphite was regenerated using acid washing (AW) and calcination (CT) methods and was then used as a Fenton-like catalyst for bisphenol A (BPA) removal. The results showed that the AW + CT process reduced defects in the graphite

structure, removed metal impurities, and promoted the formation of Mn-O bonds and oxygen-containing functional groups on the surface. These active sites accelerated the adsorption of Fe^{3+} ions and the Fe^{3+}/Fe^{2+} cycle, thereby increasing the formation of hydroxyl radicals and facilitating the oxidative degradation of BPA. Consequently, the recycled graphite exhibited superior catalytic performance compared with commercial graphite, establishing itself as a promising material for water treatment applications (Peng et al., 2025). Hao et al. (2020) investigated converting waste graphite from used lithium-ion battery anodes into an efficient heavy metal adsorbent. To achieve this, the recovered graphite was first activated via high-energy ball milling to transform it into an amorphous carbon structure. It was then coated in situ with MnO_2 to produce the AG@MnO_2 adsorbent. Ball milling increased the graphite surface area by approximately 25-fold, increasing defects and functional groups to facilitate MnO_2 loading. The resulting material was used to remove Cd(II) from an aqueous solution, and its maximum adsorption capacity was found to be 135.81 mg/g. Mechanism studies revealed that Cd(II) removal primarily occurred due to electrostatic attraction between the negatively charged MnO_2 and carbon surfaces, as well as between the Cd^{2+} ions and these surfaces. Surface complexation and ion exchange also contributed to the process. In conclusion, the study demonstrated that waste battery graphite can be converted into high-performance water treatment materials through nanostructural modifications (Hao et al., 2020). Yu et al. (2021) aimed to transform waste graphite derived from spent lithium-ion battery anodes into a valuable product by synthesising graphene oxide (GO) via a modified Hummers method. Analysis demonstrated that contaminants present on the graphite surface, such as PVDF, $LiPF_6$, LiF, Li_2CO_3 and CuO, were effectively removed during oxidation. The analyses also showed that the graphite layers had been exfoliated, increasing the surface area by 100–200 times and the interlayer spacing from approximately 0.34 nm to 0.7–0.8 nm. This resulted in a 2D graphene oxide structure. In conclusion, this study shows that waste battery graphite can be used directly to produce high-value graphene oxide, a valuable resource for energy storage, sensors, and environmental applications (Yu et al., 2021). Liu et al. (2020) first purified waste graphite obtained from used lithium-ion battery anodes using sulfuric acid to remove metal impurities and electrolyte residues. The graphite was subjected to a carbothermal structural reconstruction process involving an iron catalyst and glucose, converting it back into a high-performance anode material. This process repaired the graphite's degraded crystal structure, bringing its interlayer spacing closer to that of commercial graphite and increasing the degree of graphitisation. In electrochemical tests, the reconstructed graphite (RG) demonstrated a capacity of 427.9 mAh/g and high-rate performance after 200 cycles in lithium-ion batteries. Additionally, acid-treated graphite (AG) was used directly as an anode in sodium-ion batteries, achieving a capacity of 127 mAh/g at 50 mA/g and retaining 90.98% of its capacity after 500 cycles. In conclusion, this study has demonstrated that waste graphite can be converted back into a battery anode through nanostructural and crystallographic restoration, making it a potential high-performance energy storage material for use in both lithium and sodium-ion batteries (Liu et al., 2020). Meng et al. (2022) simultaneously recovered the cathode ($LiFePO_4$) and anode (graphite) from used $LiFePO_4$/graphite lithium-ion batteries and incorporated them into a new battery system. To achieve this, the waste batteries were first discharged and then disassembled. The $LiFePO_4$ and graphite electrode materials were then separated and ground to produce a recycled $LiFePO_4$/graphite (RLFPG) composite. This composite was then integrated into a dual-ion battery (DIB) system, acting as a cathode that can store both Li^+ and PF_6^- ions. Mechanism studies revealed that this capacity is formed through the intercalation of Li^+ ions into the $LiFePO_4$ structure at low voltages, and the intercalation of PF_6^- ions between graphite layers at high voltages. Consequently, the recycled RLFPG cathode exhibited a capacity of 117.4 mAh/g at a current density of 25 m/g and retained 78% of its capacity after 1,000 cycles. This study demonstrates that waste graphite can be utilised as both a re-anode and an active electrode material in next-generation dual-ion batteries in conjunction with $LiFePO_4$ (Meng et al., 2022).

Recent studies show that spent lithium-ion battery graphite can be reused as battery anodes and converted into graphene oxide, adsorbents, catalysts, and advanced functional carbon materials. Changes occurring at the nanoscale during these transformation processes, such as the elimination of surface defects, the reorganization of the crystal structure, the optimization of interlayer spacing, and surface functionalization, play a critical role. Therefore, nanotechnology emerges not only as a tool to improve graphite recycling performance, but also as a fundamental approach to repurposing waste

materials. With strategies such as nanostructural restoration, defect engineering, and surface modification, recycled graphite can exhibit properties comparable to, or in some cases superior to, those of the original material. These developments demonstrate that graphite recycling has evolved from a traditional recovery process to the production of high-value materials and highlight the significant opportunities that nanotechnology-supported approaches offer for sustainable energy storage systems, environmental applications, and the circular economy. Fig. 6 presents a flowchart illustrating the main steps involved in the nanostructural restoration of recycled graphite.

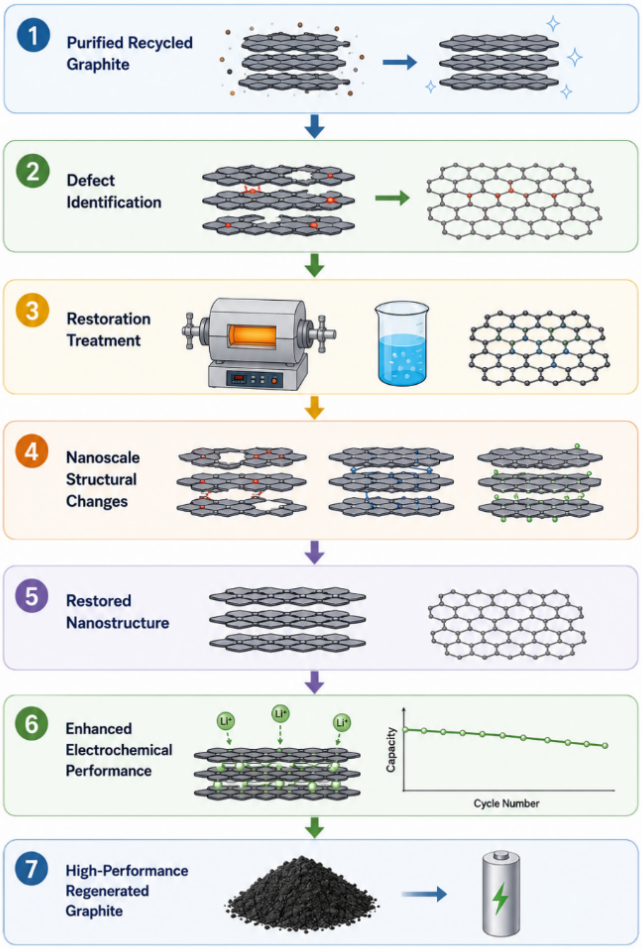

Fig. 6. Schematic representation of the nanostructural restoration of recycled graphite

Table 2 summarizes representative studies on the regeneration and reutilization of graphite recovered from spent batteries. As shown in Table 2, regenerated graphite has been successfully applied not only as an anode material in lithium-ion and dual-ion batteries but also in high-value applications including wastewater treatment, heavy metal adsorption, and catalytic degradation of organic pollutants. Depending on the regeneration strategy used, recycled graphite can show electrochemical capacities comparable to commercial graphite while maintaining excellent cycling stability and high removal efficiencies in environmental applications. These findings demonstrate that nanostructural restoration and surface modification enable spent graphite to be transformed into a multifunctional carbon material with applications extending well beyond battery recycling, thereby supporting both resource sustainability and circular economy principles.

6. Conclusions

This study comprehensively evaluated graphite's critical role in lithium-ion batteries, the properties of natural and synthetic graphite, and the importance of direct recycling in sustainable battery technologies. From a mineral processing perspective, graphite's natural hydrophobicity enables efficient recovery from black mass using flotation techniques while preserving its structural integrity.

Table 2. Performance and applications of regenerated graphite recovered from spent batteries

Source of Spent Graphite	Recycling Method	Intended Use	Capacity	Cycle Life	Efficiency	Source
Spent lithium-ion batteries	Stepwise purification and calcination (500 °C)	Anode material for lithium-ion batteries	359.40 mAh/g	100	87	(Ji et al., 2024)
Failed commercial 3.2 V, 5 Ah cylindrical (32650) LFP battery	Sonication in DI water, drying at 70 °C, and thermal treatment at 800 °C	Anode for Li-ion based all-carbon dual-ion battery (ACDIB)	250 mAh/g	300	99	(Kayakool et al., 2021)
Spent Lithium-Ion Batteries (NCM cathode residues)	Acid treatment followed by carbothermal reduction	Anode for Lithium Ion Batteries (LIBs)	427.9 mAh/g	200	79	(Liu et al., 2020)
Spent LCO batteries	Acid leaching	Anode materials in new LIBs	377.3 mAh/g	-	-	(Yu et al., 2021)
Spent Graphite/LiFePO4 batteries (Anode)	Manual separation, grinding, and mixing with cathode (concurrent reuse)	Cathode material for dual-ion batteries (DIB)	117.4 mAh/g	1000	-	(Meng et al., 2022)
Spent commercial LIB anodes	Immersion in DI water, filtration, drying (80 °C), heating under (600 °C), and hydrothermal grafting	Wastewater treatment (Pb(II), Cd(II), and Ag(I) removal)	92.35 mg/g (Pb), 23.25 mg/g (Cd), 62.7 mg/g (Ag) (Langmuir capacity	-	99.9% (Pb), 79.7% (Cd), 99.8% (Ag) removal rate	(Zhao et al., 2017)
Spent Lithium-Ion Battery (Anode)	Ultrasonication, leaching, calcination, ball milling, and coating	Adsorbent for Cadmium (Cd(II)) removal	135.81 mg/g	4	99.53% removal rate (at 0.8 g/dm^3 dose)	(Hao et al., 2020)
RGP-ARAW, RGP-BRAW, RGP-CRAW (NMC/SEI waste)	Acid washing and calcination	Fenton-like catalyst for wastewater purification (Bisphenol A removal)	-	5	-	(Peng et al., 2025)

This represents a significant advantage over conventional recycling methods, which often involve the complete decomposition of materials into their elemental forms. Various thermal, chemical, electrochemical, and hybrid methods used for cathode relithiation and graphite anode purification offer distinct advantages and limitations. However, direct recycling represents a particularly promising approach due to its lower energy consumption, reduced chemical usage, and significantly smaller

carbon footprint. By preserving the embodied energy of materials, this method minimizes environmental impact while eliminating the need for energy-intensive material re-synthesis, thereby creating substantial economic value. The high NPV and short investment payback period demonstrate the strong industrial applicability of direct recycling. Moreover, the local recovery of critical raw materials such as graphite and lithium offers strategic advantages in terms of supply security and supports the establishment of closed-loop battery production. Thus, direct recycling provides a practical pathway for retaining both the structural and economic value of spent graphite while reducing dependence on primary graphite resources.

In recent years, studies have shown that waste graphite is not only a valuable battery component that can be recovered, but also a significant carbon source that can be transformed into high-value products through nanotechnology-enabled restoration and modification strategies. Through nanoscale processes such as defect engineering, surface modification, structural restoration, and functionalisation, recycled graphite can be reused as a battery anode or converted into advanced functional products, including graphene oxide, adsorbents, and catalytic materials. The novelty of this review lies in integrating conventional graphite recovery and direct recycling with nanostructural restoration and high-value reutilization, thereby presenting graphite recycling as a transition from simple material recovery toward functional material regeneration. This perspective highlights the potential of nanotechnology not only to restore the electrochemical performance of spent graphite but also to expand its applications beyond battery reuse.

Future research should therefore focus on scalable and energy-efficient restoration processes, standardized quality criteria for recycled battery-grade graphite, and a better understanding of how defect density, interlayer structure, and surface chemistry affect long-term electrochemical performance. Integrating life-cycle assessment and techno-economic analysis will also be essential to ensure performance improvements remain environmentally and economically sustainable. Ultimately, combining direct recycling with nanostructural restoration could transform spent graphite into a strategic source of battery-grade and high-value functional carbon materials, supporting sustainable energy storage and a closed-loop battery economy.

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