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Review Article

Toward sustainable graphite anode recycling: From degradation understanding to targeted regeneration

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ABSTRACT

The rapid proliferation of spent lithium-ion batteries has created an urgent need for efficient recycling of graphite anodes, yet industrial adoption remains constrained by economic and technical barriers. This review systematically examines the underlying causes of this stagnation by analyzing current industrial practices, graphite degradation mechanisms, and academic recycling strategies. Analysis of the industrial landscape reveals that prevailing recycling processes roughly adapt from primary material production, leading to an imbalanced industry structure and a fundamental mismatch between industry graphite recycling and market demands. Further investigation into degradation mechanisms demonstrates that the decline in electrochemical performance is predominantly governed by surface-level deterioration, revealing the mismatch mechanism. In response to the mismatch, academic advances have developed targeted regeneration strategies that directly address the specific failure modes of spent graphite, thereby offering viable pathways to resolve the industrial mismatch. Therefore, by integrating insights from both industrial practice and academic research, this review delineates a clear pathway forward that graphite recycling must undergo a paradigm shift from rough reconstruction toward precision repair.

KEYWORDS

graphite anode, lithium-ion battery recycling, degradation mechanisms, battery sustainability, circular economy, surface solid electrolyte interphase (SEI)

1 Introduction

Graphite, composed of stacked graphene layers with carbon atomic sp^2 hybridization, exhibits a regular layer structure with a layer spacing of 3.35 Å [1–3]. This structure enables fast electron transfer in plane and rapid thermal spread in the form of phonons, endowing graphite with excellent thermal and electrical properties. Additionally, the regular layer structure and appropriate layer spacing provide sites for lithium-ion diffusion and intercalation to form LiC_6, facilitating the industrial development of lithium-ion batteries (Fig. 1(a)) [4, 5]. Not only in the battery industry, but also in various fields, the excellent properties of graphite material promote its wide application [6]. Therefore, the emerging material demands establish a large graphite market, the size of which is projected to reach $23.87 billion by 2033 with a compound growth rate of 7.8% from 2026 to 2033 [7]. Especially, the demand for battery-grade graphite with high purity is set to triple to 2.7 million tons by 2030 [8, 9].

To satisfy the increased demand for graphite material, technologies and amount of capital are input into the graphite production, separating the production routes to purification of natural graphite and synthetic graphite [10–13]. During the natural graphite route, the mined graphite ore from open-pit mining or underground mining is first purified by a flotation process, followed by chemical purification to separate the graphite particles from the other minerals [14]. Then, the floated graphite minerals are crushed, milled and sieved, removing the larger graphite particles. Further removal of impurities is required by chemical refining, which commonly involves acid washing and heat treatment [15], facilitating their applications in battery materials, lubricants, refractories, lead-acid batteries, and many others. The artificial graphite route takes petroleum coke (or coal

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tar, asphalt, etc.) as raw material for the following carbonization and graphitization [16]. After mixing the powdered petroleum coke with additives and molding or extruding it into the desired shape, high pressure is required to ensure the material density and stability [17]. Then, the feedstock enters a high-temperature carbonization and graphitization. The carbonized material is graphitized at higher temperatures (2500–3000 °C) to promote the rearrangement of the carbon atoms in layer structure, for following sieved, cutting, and other post-processing to obtain a graphite product that meets specifications. Stimulated by the demands in battery and other fields [18–22], the production of graphite material, both in natural and synthetic graphite, exhibits a rapid output growth. The global graphite production reached 1.73 million tons in 2024, with China being the largest producer of 1.27 million tons [23]. Although short-term supply and demand contradictions could be alleviated by expanding the scale of extraction, the geographical concentration of natural graphite resources and the high energy consumption/pollution of synthetic graphite make production models that rely on unsustainable resources in the long run. Therefore, there is an urgent need to develop graphite recycling to solve this dilemma [24–34].

Herein, we reveal the mismatch between industry graphite recycling and market demand through the analysis of industrial status, degradation mechanisms and academic recycling research. The industrial status reveals an industrial pattern characterized by resources and supply chains with rudimentary recycling processes derived from graphite production, highlighting the effects and consequences of the mismatch. To understand the reason for the mismatch, further analysis of the degradation mechanism reveals that the surface degradation dominates the attenuation of electrochemical properties, requiring targeted surface repair. Based on this understanding, advanced recycling methods validate the feasibility of repair through accurately targeting the degradation mechanisms of spent graphite, indicating the possible solutions for the mismatch. Therefore, the combination of industry and academic analysis reveals the route that graphite recycling should be transformed from rough reconstruction into targeted repair.

2 Industrialization status of graphite recycling

Lithium-ion batteries (LIBs) have gained widespread adoption across energy storage systems, particularly in electric vehicles (EVs), fueling exponential growth in LIB production. The global LIB shipments surged to 1545 GWh in 2024 (Fig. 1(b)), representing a remarkable 28.5% year-on-year increase [35]. However, the inherent limited lifespan leads to the retirement of batteries after 5–8 years [25]. The impending tidal wave of retired LIBs (Fig. 1(c)), projected to create a $13.5 billion market by 2030 [26], mandates comprehensive graphite recycling industrially. In this section, the industrial graphite recycling is discussed and analyzed from the recycling output capacity, market and technology supporting.

Surging demand for lithium-ion batteries and the retirement amount stimulate the growth of the graphite recycling market. With the coming of the tidal wave of spent batteries (Power batteries installed in 2017–2018), the global spent battery materials are anticipated to reach 1.4 million tons by 2030 [36, 37]. The research organization estimates that the total volume of spent batteries in China would climb toward 1 million tons by 2030 [38]. In these batteries, the graphite material accounts for ~15% battery mass, which could be estimated as the mass of 0.21 million tons of global spent graphite. This growing retirement volume establishes a market size of graphite recycling to $1.42 billion by 2024, and it is anticipated to reach $2.98 billion by 2033 with a CAGR of 8.7% [39]. If recycling the overall spent graphite, its output would occupy ~12.5% of the global graphite market by 2033 (~$23.8 billion), exhibiting a large gap between supply and demand. Although the market size is growing, the growth rate is slower than that of the cathode material due to the existence of valuable metals (Fig. 1(d)), indicating the presence of significant inhibition factors.

The graphite recycling markets are categorized by region, including the Asia Pacific, North America, Europe, the Middle East, Africa and South America [7, 40]. During these regions, the Asia Pacific dominates the recycling market of graphite due to excellent battery manufacturing and large graphite output. Especially in China, Japan and Republic of Korea, which exhibit the leadership in battery, electronic and vehicle production. Therefore, the Asia Pacific region demonstrates the largest market with a proportion of 52% (~$0.74 billion) of the world, while it is estimated to maintain its leadership with a CAGR of 9.2% [39]. North America occupied 21% of the global recycling market of graphite by 2024, reaching ~$0.3 billion. Meanwhile, benefited by the policy support and technology advances, the market size would continuously expand. With the management of environmental policies in Europe and mature industrial techniques, Europe occupies ~19% of the graphite recycling market, resulting from the investment in battery recycling and sustainable manufacturing. Although the market is relatively smaller than other regions, it is expected to grow with the improvement of industrialization and the cognitive transformation of sustainable materials. Then, the differences between the market size and industrialization should be further understood by analyzing the industrial structure. The companies are categorized based on their role in the industry chain, containing graphite production enterprises, material recycling enterprises and crossover enterprises with advanced technologies. For example, GrafTech, Superior Graphite, SGL Carbon, RS Mines, EcoGraf, Ceylon Graphite Corp., Graphite India Limited, Imerys Graphite & Carbon, Talga Group, Shanshan Technology and Nippon Carbon extend their material production or mine development business of graphite to spent graphite recycling [39–41]. GEM, Brunp, Carbide and Semco Carbon, etc., enterprises mainly focus on material recycling and expanded graphite recycling. The distribution of enterprises and their roles in the industry chain were collected and listed in Table 1. In these companies, the graphite recycling is still concentrated on the mining and material process companies, whose proportion surpasses that of the recycling companies. For material recycling, companies mainly rely on the existing battery recycling chain, such as GEM and Brunp, etc., or advanced technologies, such as Altilium and Tozero, etc., forming an individual industry ecosystem. However, fewer companies recycle spent graphite to make battery anode and close the loop of the battery chain, isolating the recycling and battery application. Besides, the number of enterprises and the production capacity of graphite recycling are still less than those of material production enterprises. Therefore, the differentiation of regional patterns reflects the strategic differences in the graphite recycling industry of various countries.

To further reveal the industrialization status of graphite recycling, the recycling processes of typical enterprises were analyzed. During battery recycling, discharge treatment is the first step in recycling battery materials, including salt solution

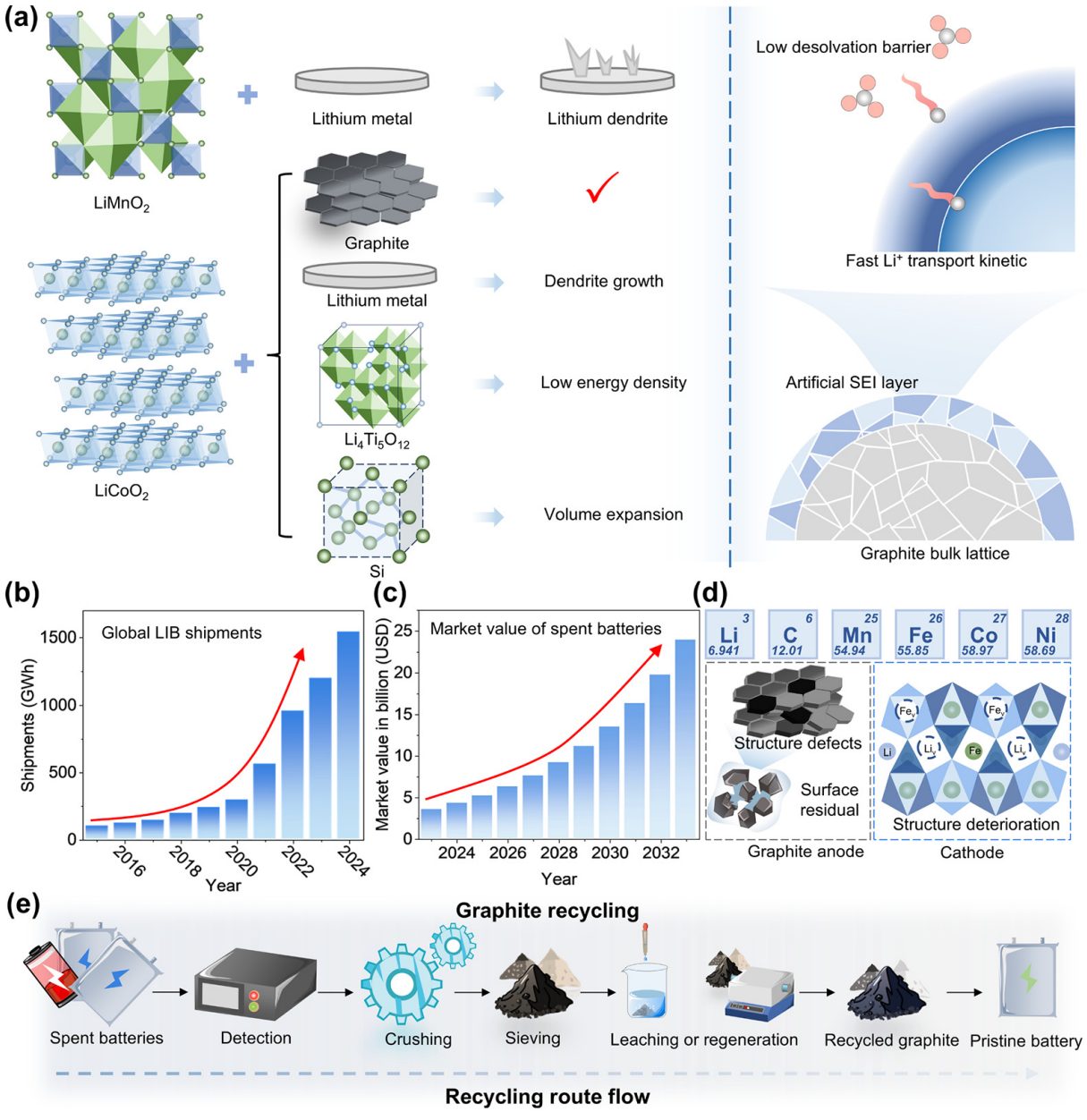

Figure 1 Graphite anodes face significant recycling challenges driven by the large-scale production and retirement of lithium-ion batteries. (a) The evolution history of the lithium-ion battery and graphite anode. (b) Global shipment of lithium-ion batteries from 2015 to 2024. Based on data from Ref. [24]. (c) Market value growth of the global battery recycling industry. Based on data from Ref. [26]. (d) Schematic structure of a spent battery, highlighting metallic components from cathodes and carbonaceous materials from anodes and additives. (e) The typical recycling route of the spent graphite anode, in which the spent batteries are detected, crushed and sieved to obtain the graphite material for subsequent impurity removal and regeneration.

discharge, solid conductor discharge, low-temperature freezing chemical discharge, etc. [42]. In the industrial sector, companies commonly discharge batteries by immersing them in a salt solution (most commonly NaCl or other inorganic salts [43]) with a mass fraction of 1%–5%. This method could process discarded batteries on a large-scale. However, high salt concentration or prolonged immersion time leads to corrosion of the metal casings and electrolyte leakage, resulting in severe wastewater and electrode contamination. Differently, companies Brunp, Umicore, and Toxco [44–46] utilize liquid nitrogen to rapidly cool batteries to ultra-low temperatures (lower than -100 °C), effectively reducing internal reactivity and enabling safe mechanical crushing without prior discharging. Brunp and the EKATO Group [47, 48] conduct battery crushing in a vacuum environment. By eliminating oxygen, this technology prevents exothermic oxidation reactions of electrolyte residues or lithium metal. At the same time, the unique properties of the vacuum environment are utilized to optimize the crushing effect, improving the recovery rate of metals and active materials. However, these methods impose relatively high requirements on the equipment, rendering them less suitable for large-scale industrial applications.

Umicore and BARTEC [49] in Europe use the proprietary VAL'EAS pyrometallurgy process to recycle lithium-ion batteries, producing cobalt or nickel alloys, while graphite and organic solvents are utilized as fuel to release energy. This method is capable of processing large quantities of spent lithium batteries, and Umicore's Hoboken plant in Antwerp, Belgium, processed 7000 tons of retired batteries by 2022 [50]. With the increase in the need to close the loop, some enterprises are investing in the graphite recycling industry. A prime example is the collaboration

Table 1 Main companies and distribution of global graphite recycling

Number	Company	Region	Industry chain situation
Asia Pacific			
1	Fangda Carbon New Material Co., Ltd.	China	Material production
2	Shanshan Technology	China	Material production
3	Jiangxi Zichen Technology	China	Material production
4	GEM	China	Material recycling
5	Brunp Recycling	China	Material recycling
6	Toyo Tanso Co. Ltd.	Japan	Material production
7	Nippon Carbon Co. Ltd.	Japan	Material production
8	SEC Carbon Ltd.	Japan	Material production
9	Tokai Carbon Co., Ltd.	Japan	Material production
10	Resonac	Japan	Material production
11	Dowa Holdings	Japan	Material recycling
12	Graphite India Limited	India	Material production
13	Epsilon Carbon	India	Material production
14	HEG Limited	India	Material production
North America			
15	GrafTech International	America	Material production
16	Superior Graphite	America	Material production
17	Carbon Activated Corporation	America	Material production
18	Asbury Carbons	America	Material production
19	Carbide Recycling Company	America	Material recycling
20	X-BATT	America	Material recycling
21	Semco Carbon	America	Material recycling
22	Weaver Industries, Inc.	America	Material recycling
23	Ascend Elements, Inc.	America	Material recycling
24	ReElement Technologies	America	Material recycling
25	Ceylon Graphite Corp.	Canada	Mining Industry
26	Focus Graphite Inc.	Canada	Mining Industry
27	Northern Graphite Corporation	Canada	Mining Industry
28	Mason Graphite Inc.	Canada	Mining Industry
29	Lab4 Inc.	Canada	Material recycling
Europe			
30	SGL Carbon	Germany	Material production
31	GAB Neumann GmbH	Germany	Material production
32	Duesenfeld GmbH	Germany	Material recycling
33	tozero	Germany	Material recycling
34	Imerys Graphite & Carbon	France	Material production
35	Mersen	France	Material production
36	Coidan Graphite	Britain	Material production/recycling
37	Altilium	Britain	Material recycling
38	Elkem ASA	Norway	Material production
39	Umicore	Belgium	Material production
Oceania			
40	EcoGraf Limited	Australia	Material production/recycling
41	Triton Minerals Ltd.	Australia	Mining Industry
42	Syrah Resources	Australia	Mining Industry
43	Talga Group	Sweden/Australia	Mining Industry
Other			
44	RS Mines	Sri Lanka	Mining Industry

of the Chinese firm GEM with China Minmetals Corporation [51]. The partners have established an advanced battery recycling system for retired power cells through synergistic integration of physical crushing and hydrometallurgical processes. Building on this technological foundation, they have successfully industrialized graphene-enhanced lithium iron phosphate (LFP) materials, creating a closed-loop industrial chain that spans from spent lithium battery recovery to high-purity graphite production and lithium anode material synthesis. Brunp in China could obtain high-performance recycled graphite (Impurities<100 ppm) by sequential precision filtration, spray drying, and microwave heating of a dispersion containing used high-purity graphite powder [52]. Britain's graphite company Altilium uses proprietary EcoCathode™ technology (Chemical leaching and high-temperature treatment) to recover over 97% lithium and 99% graphite from LFP batteries and reuse them in new batteries straight and the Teesside recycling plant could recycle the graphite, which is enough to meet over 20% of UK demand by 2030 [53, 54]. Besides, Altilium and Talga have built a partnership to recycle graphite anode, and Talga would use chemical purification methods to recycle graphite anode with excellent performance and physicochemical properties [55]. Similarly, Australian graphite developer EcoGraf™ proposed an HF-free strategy to recycle spent graphite with a purification of 99.95%, which reduces carbon emissions and supports the circular economy. Nowadays, this enterprise has established several recycling plants alongside purification facilities [56]. In America, Ascend Elements and Koura Global focused on battery recycling, and they received a USD 125 million cost-sharing grant from the government to recycle graphite. Ascend Elements proposed a Hydro-to-Anode technology combined with purification to recycle graphite with excellent performance and low waste emission [57, 58]. Redwood Materials in America acquires retired power batteries from its partners Tesla, Ford, Toyota, etc., and also converts them into high-quality battery materials and sells them to battery manufacturers, of which 95%–98% of the effective materials in the retired batteries would return to the industry chain [59]. Based on the conclusion of the above recycling status, several companies are laying out graphite anode recycling, and graphite provides energy in other material recycling processes (Fig. 1(e)). Although hydrometallurgy leaches out the residual metals in graphite, further long-term high-temperature heating is required to recover the graphite properties.

Therefore, the graphite recycling industry is primarily located in the leading regions of the battery industry and is derived from graphite production enterprises that can apply their production processes to graphite recycling. Although battery recycling enterprises have paid some attention to graphite recycling, graphite plays a role as a byproduct or raw material in cathode recycling, significantly reducing the application value of regenerated graphite. The uneven distribution, disequilibrium development, and imbalanced supply and demand demonstrate the mismatch between the graphite recycling industry and graphite demand.

3 Degradation mechanism of the graphite anode

The current industrial methods (high-temperature treatment, acid leaching) try to reconstruct the whole particle, while graphite degradation mainly occurs in surface SEI and near-surface defects with basically intact bulk [60]. This overtreatment has led to high energy consumption, low efficiency and poor economy. To break this dilemma, it is urgent for the academic community to start with the degradation mechanism and provide theoretical guidance and technical solutions for industrial precise repair [61].

During battery cycling, lithium ions are deintercalated from the cathode to the bulk electrolyte, forming a Li^+-solvent structure with an anion that transfers the lithium ions in the inner electrical field [62]. When the Li^+-solvent ion pair transfers through the electrolyte to the anode surface, the dissolved lithium ion crosses the surface of the preformed solid-electrolyte interphase (SEI), diffusing into the interphase to the graphite particle surface. Then, driven by the electrode potential, the ions further diffuse to the graphite edge planes and form a LiC_x structure with the continuous intercalation based on the stage mechanism [63]. Therefore, the intercalation/deintercalation process of lithium ions experiences interface reaction and bulk diffusion driven by potential, where the material properties limit the intercalation kinetics in the process. For example, desolvation rate, SEI conductivity, and intercalation kinetics in the graphite layer affect the ion transfer and intercalation process [5, 64]. When the ionic deintercalation/intercalation could not couple with electrons, ion aggregation results in electronic field polarization. Therefore, the various utilization situations may degrade the battery through the irreversible side reaction and structure damage of battery materials after long-term cycling, inhibiting the electrochemical properties of the graphite anode [65]. The degradation mechanism of spent graphite anode needs to be paid attention to, establishing the relationship between graphite degradation mechanisms and electrochemical performance.

One of the primary degradation phenomena is the overgrowth of the uneven SEI layer, which has been recognized as the main reason for lithium loss [66]. The SEI layer forms at the anode surface during the charge with the electrolyte reduction, separating the electrolyte and graphite anode to work the battery below the reduction potential of the electrolyte and transfer the lithium ions. Therefore, this layer gradually evolves from its initial state with the time and cycling extension, resulting in an uneven and thick SEI layer. Continuous SEI growth during cycling increases the ion transport impedance [67], further improving the electrical field polarization while consuming active lithium (Fig. 2(b)). When increasing the Li^+ gradient to a high concentration which surpasses 0.077 $mol \cdot cm^{-3}$ near the graphite surface, the aggravated polarization induces a lower potential to below the deposition potential of lithium [68]. This high concentration and low potential lead to the lithium deposition and dendrite formation on the graphite surface. Gradually formed lithium dendrite not only exacerbates SEI growth on the dendrite surface and exposed graphite surface but also raises safety risks of separator impalement. Besides, electrolyte reaction and SEI growth on some separated lithium dendrite surfaces could isolate the lithium and graphite anode due to the electronic insulation of the SEI layer, forming the irreversible dead lithium [69]. Besides the byproducts, graphite bulk also suffers from property deterioration during the electrochemical cycling. In the lithium intercalation process, Li^+-solvent configuration, SEI layer and solvent selection determine the desolvation process and behavior. An inadequate desolvation process results in the transfer of the solvated lithium ion across the SEI layer to the graphite layer. This process leads to the exfoliation of the surface layer of graphite and bulk lattice defects, causing the capacity decay. Besides, irreversible intercalation of lithium ions in the graphite structure also damages the lattice structure, resulting in the loss of active mass and capacity loss [70]. Therefore,

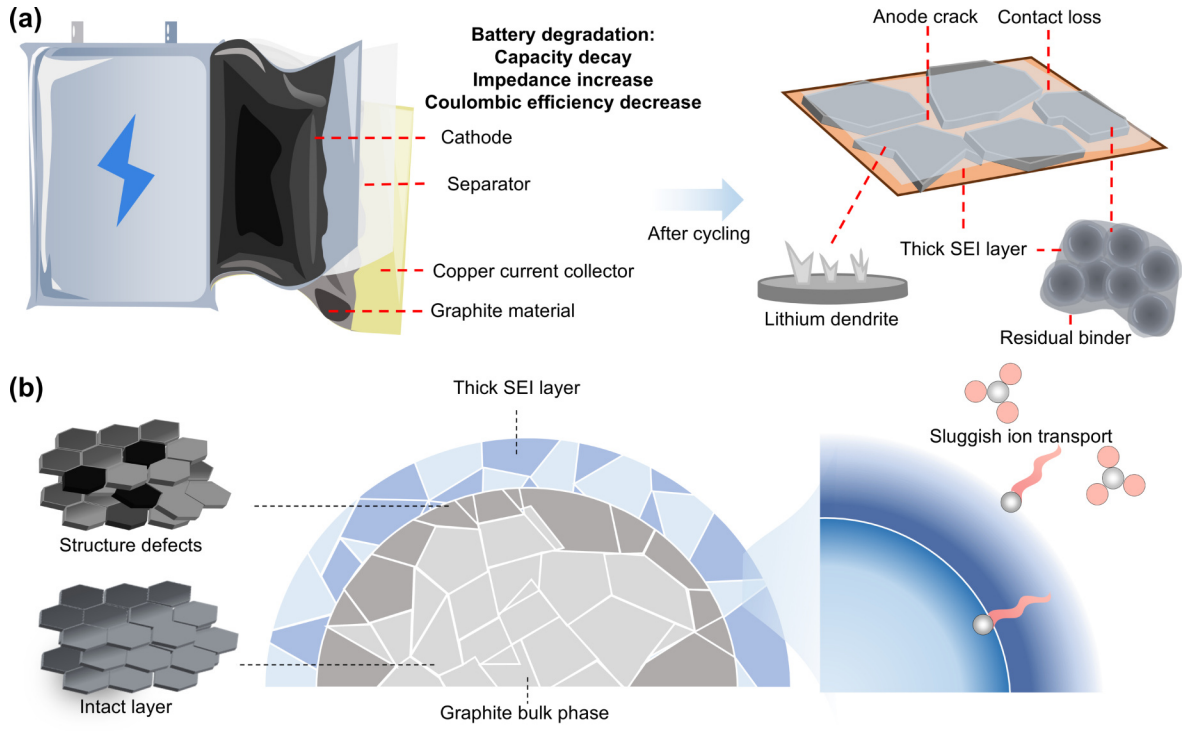

Figure 2 Degradation mechanisms of graphite anodes in spent lithium-ion batteries. (a) Schematic illustration of the dominant degradation mechanisms in spent graphite anodes. During repeated charge–discharge cycles, the graphite anode undergoes several interrelated degradation processes. (b) Schematic illustration of microscopic degradation mechanisms and their effects on ionic transport. At the nanoscale, the degradation is predominantly localized at the particle surface and near-surface regions. The thickening SEI layer blocks lithium-ion diffusion pathways and increases charge-transfer resistance. Near-surface lattice distortions, including layer expansion, edge dislocations, and point defects, further impede ion intercalation.

repeated deintercalation/intercalation of lithium ions and inadequate desolvation induce volume change of the graphite lattice, causing residual mechanical stresses and exhibiting lattice expansion, exfoliation, and defect formation.

In summary, the graphite degradation is correlated with the attenuation of electrochemical performance, focusing on the surface or near-surface region, which limits the electrochemical reaction kinetics. For example, SEI growth, lithium dendrite and graphite exfoliation mainly concentrate at the graphite particle surface. Besides, the excessive SEI growth has been recognized as the main reason for lithium loss, resulting in the battery capacity decay. The thick and uneven SEI layer also contributes to the increased impedance of the battery, limiting interface reaction kinetics [71]. These not only inhibit the cycling stability of the battery but also deteriorate the rate capability for fast charging. Although the bulk phase of graphite exhibits some defects, such as lattice expansion and dislocation, the graphite lattice still provides sufficient sites for lithium intercalation, while the simple surface repair could recover the performance of graphite.

4 Academic research on the spent graphite recycling

To resolve the industry mismatch, advanced targeted repair of spent graphite has been developed based on the understanding of degradation mechanisms. The repair process could be separated into surface binder removal, surface impurity removal and lattice repair, which mainly focuses on the regeneration of spent graphite for battery application.

4.1 Surface binder removal of spent graphite

The recycling strategies mainly involve breaking down batteries and recovering individual components. Effective material separation is critical to avoid impurity detriment on the electrochemical performance of regenerated electrodes [72, 73]. After separation, electrode materials differ from commercial ones in the presence of binders, electrolyte residues, and degradation-induced defects, necessitating additional regeneration steps [74, 75].

Removing organic binders is a critical step to liberate graphite from the copper current collector [76], where thermal decomposition is the predominant strategy for binder removal. For example, Chen *et al.* [77] demonstrated air pyrolysis between 300–900 °C, effectively burning off binders, albeit generating toxic emissions such as dioxins and fluorinated gases. To mitigate the environmental impacts of exhausted gases, Zhang *et al.* [78] explored anaerobic pyrolysis under nitrogen or vacuum, enabling binder decomposition at 500 °C into collectible oils and gases (Fig. 3(a)), improving graphite recovery from 88.08% to 99.60% with a smoother surface (Fig. 3(b)). Besides the removal, controlled thermal heating could transform the residual organic into the carbon layer. For example, the carbonization of surface binder was explored to eliminate the surface residual binder while forming a carbon coating on the graphite surface to improve the conductivity [79–82]. Other studies propose lowering pyrolysis temperatures or simultaneously treating collectors and anodes [83–88]. Although these methods reduce energy demands, they still face environmental concerns, necessitating advanced processing equipment.

Alternatively, eco-friendly binders, such as water-soluble CMC/SBR, enable binder removal using simple water washing, eliminating the need for high temperatures and reducing greenhouse gas emissions. Subsequently, Zhou *et al.* [89] introduced a water-assisted separation technique, leveraging hydrogen gas generated by reactions between water and lithium

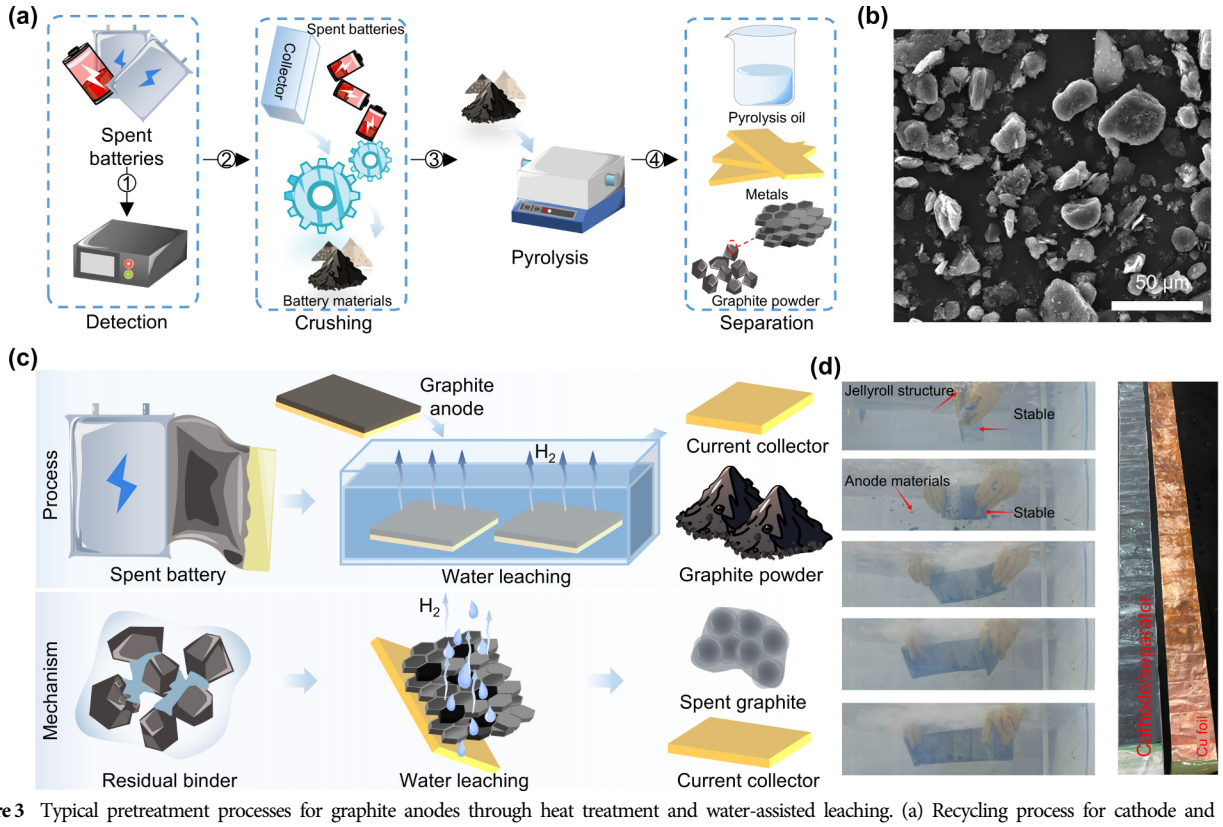

Figure 3 Typical pretreatment processes for graphite anodes through heat treatment and water-assisted leaching. (a) Recycling process for cathode and anode materials from spent LIBs, where the binder is pyrolyzed through heat treatment. (b) SEM image of pretreated graphite anode material. (b) Reproduced with permission from Ref. [78], © 2020 American Chemical Society. (c) Schematic illustration of the changes occurring in the opened jelly-roll structure. (d) Unrolling the jelly roll underwater to isolate reactive materials from oxygen and control the reaction between lithiated graphite and water. (d) Reproduced with permission from Ref. [89], © 2021 Elsevier B.V.

residues (Figs. 3(c) and 3(d)). Residual impurities were removed through mild heating, facilitating lattice repair and restoring electrochemical performance. Besides, due to the water-solubility, ultrasonic or hydrothermal methods were proposed to completely separate the binder from the graphite surface, facilitating the further impurity removal and lattice repair [90, 91]. Overall, pretreatment advances, particularly binder removal innovations, lay the foundation for efficient graphite recycling.

4.2 Surface impurity removal of spent graphite

After the surface binder removal, further regeneration pays attention to the graphite degradation mechanisms during cycling, such as SEI growth, lithium plating, volume expansion and local stress-induced damage, which deteriorate the ionic conductivity and structural stability [92].

The impurity removal on graphite anode typically uses acids [93–98], alkalis [99, 100], or other solutions [88, 101–103] to leach metallic impurities from the electrode into the solution, obtaining high-quality graphite and metal resources. The selection of the leaching solution depends on factors such as the target metal, graphite composition, and required extraction efficiency. Inorganic acids are commonly used to address insoluble species like LiF and $ROCO_2Li$. Ma *et al.* [98] proposed a sintering-leaching approach using H_2SO_4/H_2O_2 mixtures and NaOH activation, restoring the graphite structure and achieving discharge capacities comparable to commercial graphite. The combination of acid leaching and calcination was also used to remove the impurities and repair the structure. For example, after the H_2SO_4/H_2O_2 leaching, the low-temperature calcination replaced the alkali activation to repair the graphite structure [104]. Besides the graphite recycling, the lithium in the spent graphite material could be recycled in the leaching process. Guo *et al.* [105] demonstrated HCl leaching (optimal at 80 °C, 3 M HCl, solid/liquid ratio 1:50), achieving 99.4% lithium extraction. Arshad *et al.* [106] achieved 96.1% lithium extraction and structural repair using oxalic acid, enhancing the graphite cycling stability. Ammonium persulfate $(NH_4)_2S_2O_8$ has also emerged as a selective leaching agent, simultaneously enabling impurity removal and lattice expansion [107]. The regenerated graphite exhibited a high reversible capacity of 330.2 mAh·g^{-1} after 500 cycles at 0.3 A·g^{-1}, comparable to commercial graphite. Novel methods such as low-temperature fluorination and alkaline roasting enhanced graphite purity (serial number 1 in Fig. 4) [48, 49], achieving near-commercial electrochemical performance. Yang *et al.* [96] employed a two-stage calcination-leaching process, minimizing wastewater while extracting nearly 100% of metals and lithium. Besides, the roasting methods were also proposed to reduce the impurities for further separation [108, 109]. These initial acid/alkali strategies aim to completely remove the impurities in spent graphite, ignoring the impurity distribution and the environmental impact of strong acid/alkali. With further research on surface SEI components, the purification employs water washing to effectively remove the soluble lithium compounds like CH_3OLi and Li_2O while preserving the graphite lattice structure. However, the residual lithium salts and metals, such as Li_3PO_4 and iron, remain in treated graphite and require further removal after simple water treatment.

With the deep research of surface degradation mechanisms, upcycling with surface modification is widely applied in the graphite recycling process. To further improve the electrochemical

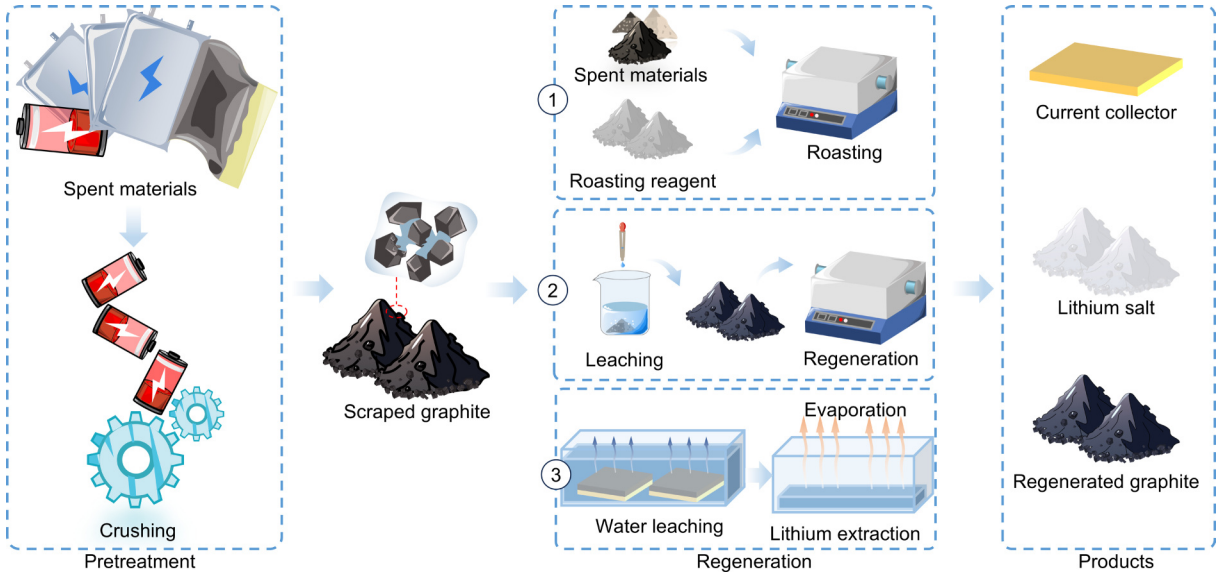

Figure 4 Schematic illustration of graphite anode regeneration to obtain current collectors, lithium salt, and regenerated graphite. Process 1. The reagent-assisted roasting for impurity removal in graphite anode. Process 2. The acid/water leaching and calcination regeneration of the graphite anode. Process 3. Water leaching to extract lithium salt and recycling the graphite anode.

properties of graphite, a calcium silicate coating layer facilitates the ion transport while protecting the graphite surface from dendrite formation and structure damage [110]. Besides the intrinsic ion-conductive SEI layer, the P or F element was introduced to the coating layer to promote the growth of Li_3P or LiF component in the SEI layer [111, 112]. For example, Zhou *et al.* [112] utilized residual Li_2CO_3 on the spent graphite surface to react with NH_4F solution, reconstructing the LiF-rich interphase with lower interfacial resistance and fast ion transport. This strategy recovered the graphite capacity and recycling stability with 303.9 mAh·g^{-1} at 0.5 C and a capacity retention of 92.3% after 500 cycles, while reducing the recycling cost of 78% compared with conventional methods.

Although different acids or auxiliary reagents leach the impurities (metals, lithium compounds, and electrolytes) in spent graphite, extensive use of acids and other reagents causes the treatment of wastewater, and post-processing, such as high-temperature sintering, is still required to address defects and crystal changes in leached graphite for reuse in LIBs. Therefore, some advanced heating methods with impurity vaporization were proposed to simultaneously remove the impurities and repair the graphite lattice, which was discussed in the section on emerging regeneration technologies [81, 82, 113, 114].

4.3 Bulk lattice repair of spent graphite

Degradation induces interlayer expansion, lattice disorder, and defect generation, necessitating secondary graphitization processes for regeneration. During the graphitization, high-temperature treatments (>3000 °C) rearrange carbon atoms to restore graphitic order, repairing the lattice defects for lithium-ion intercalation and storage.

To investigate the lattice reconstruction during the sintering process, Yu *et al.* [115] employed semi-*in-situ* X-ray diffraction, electron microscopy, and Raman spectroscopy. The study revealed that high-temperature incineration could rearrange the atomic and transform structure. Heating at 3000 °C for 6 hours was found to be the optimal condition for achieving the highest degree of graphitization and eliminating internal stress. The regenerated graphite anode exhibited a high specific capacity of 352 mAh·g^{-1}, with 97.3% capacity retention after 1000 cycles. Additionally, the pilot device for this process was able to regenerate graphite with a stable graphitization degree in 100 tests. However, the immense energy cost limits scalability. Alternative strategies integrate acid washing, catalytic graphitization (e.g., Fe- or Co-catalyzed) [116–119], or combined sintering-leaching processes at lower temperatures (900–1500 °C) [120], successfully repairing the structure with reduced environmental impact (serial number 2 in Fig. 4). To reduce the graphitization temperature, a ferrocene catalyst was used to promote structural graphitization at 700 °C [121], recovering the initial charge capacity of graphite to 359.8 mAh·g^{-1} at a 0.05 C. Notably, catalytic processes facilitate lattice reorganization but require cost-effective, scalable catalyst systems.

Some researchers employ more advanced methods to upgrade the damaged crystal lattice to a modified structure with improved ion transport kinetics. For example, the spent graphite was upcycled to a structure with rich pores at the sub-surface, which facilitates the diffusion/surface reaction kinetics for rapid ion intercalation and diffusion [122, 123]. Therefore, the upcycled graphite delivered improved rate capability and cycling stability at high rates, exhibiting 300 mAh·g^{-1} after 500 cycles at 5 C [123]. Similarly, the molten salt process or atom doping was also applied to modulate the graphite lattice structure for ion intercalation and diffusion [124, 125]. These strategies not only repair the graphite structure to its initial state but also improve the intrinsic diffusion kinetics for high-rate applications.

Spent graphite also finds secondary applications. Li *et al.* [126] utilized lithium residues in the graphite lattice to synthesize organolithium compounds (serial number 3 in Fig. 4), adding significant economic value for the precursor of material synthesis. For example, the organolithium derived from spent graphite could be synthesized as a catalyst for the PET degradation, realizing the cross-sector battery-plastic co-upcycling [127]. Ma *et al.* [101] used hydrogen generation during lithium extraction to extract the residual lithium while generating few-layer graphene flakes, enhancing the rate capabilities of regenerated graphite. Based on the property of energy barrier mitigation of delithiation, the few-layer graphene flakes were used to catalyze the delithiation of

Li_5FeO_4 on a prelithiation separator, which balances the cycling stability and energy density of the battery, realizing a power output of 1159.8 W·kg^{-1} [128]. Besides the application of lithium-ion batteries, Wu *et al.* [129] successfully reused regenerated graphite in Na and K ion batteries with a simple pyrolysis process, achieving superior performance to virgin graphite. To facilitate the ion intercalation kinetics, the carbon nanocage was fabricated through a laser process, enabling the K ion storage in regenerated graphite with 85% capacity retention after 600 cycles and the Li^+ storage with a capacity of 557.6 mAh·g^{-1}. Similarly, the graphite oxide materials could be synthesized from spent graphite and applied as an adsorbent in the wastewater process [130]. Therefore, the graphite lattice repair decides the capacity recovery, while the lattice modulation could enable the graphite for applications such as catalysts, adsorbents, or Na/K ion batteries [131]. These recycling strategies convert the simple regeneration of graphite anode in batteries to other application fields with a high added value.

4.4 Emerging regeneration technologies

Transient heating techniques such as flash Joule heating [81] (Fig. 5(a)) and rolling Joule heating [80, 82] (Figs. 5(b) and 5(c)) have been introduced to reduce energy consumption and processing time. The Joule-heated graphite preserved an initial layered graphite structure [81]. The SEI was decomposed during the Joule heating effect and simultaneously produced a carbon layer coated on the surface of the graphite. The flash-recycled anode, with a high purity of 99.5%, delivers a recovered specific capacity of 351.0 mAh·g^{-1} at 0.2 C. The cradle-to-gate life cycle assessment results reveal that the flash Joule heating method offers significant advantages in terms of energy and water consumption reduction, as well as greenhouse gas emissions. Ultimately, the continuous device and strategy were proposed to pretreat and regenerate the graphite anode. For the pretreatment of the spent electrode sheet, the rolled-over heating delivered by the Joule-heating device pyrolyzed the binder rapidly at 1500 °C and demonstrated a recycling velocity of 5 miles per minute. The recycling efficiency of the current collector and graphite material could reach 99% and 98%, respectively, while the energy input and process time were also significantly reduced. For the regeneration of spent graphite particles, the Joule-heating device was designed in a slope form that could facilitate the roll heating of graphite particles on a carbon heater [80]. After 0.1 s heating, the impurities on the graphite surface were removed and the graphitization degree was improved, delivering a capacity of 320 mAh·g^{-1} at 1 C with a capacity retention of 96% after 500 cycles. Similarly, as the fourth matter, the plasma could present a high temperature for regeneration (Fig. 5(d)) [132–134]. Xu *et al.* [71] treated the spent graphite anodes in an ethanol-water solution through ultrasonic cleaning to leach out surface impurities. After separating the graphite suspension, the leached graphite anode was treated in a nitrogen atmosphere using graphite self-induced microwave plasma for 30 seconds. The strong induced electric field generated by the plasma rapidly heated the graphite surface, removing the SEI layer while simultaneously repairing the crystal structure through the self-induced microwave plasma. The regenerated graphite obtained in this process exhibited a specific capacity of 352.2 mAh·g^{-1} at a 0.2 C. Besides, the rate capability and cycling stability were improved to the level of commercial ones, which was higher than that of spent graphite.

In summary, with a deep understanding of degradation mechanisms, the repair strategies prefer simple and targeted regeneration. Initially, long-term heating or acid leaching is widely used to remove the metal impurities and re-graphitize the whole particles of spent graphite. These methods not only consume extensive energy and chemical reagents but also produce waste gases or water, causing environmental issues and cost enhancement. After further research on degradation mechanisms and the development of transient heating technologies, the repair region gradually focuses on the near-surface of graphite particles,

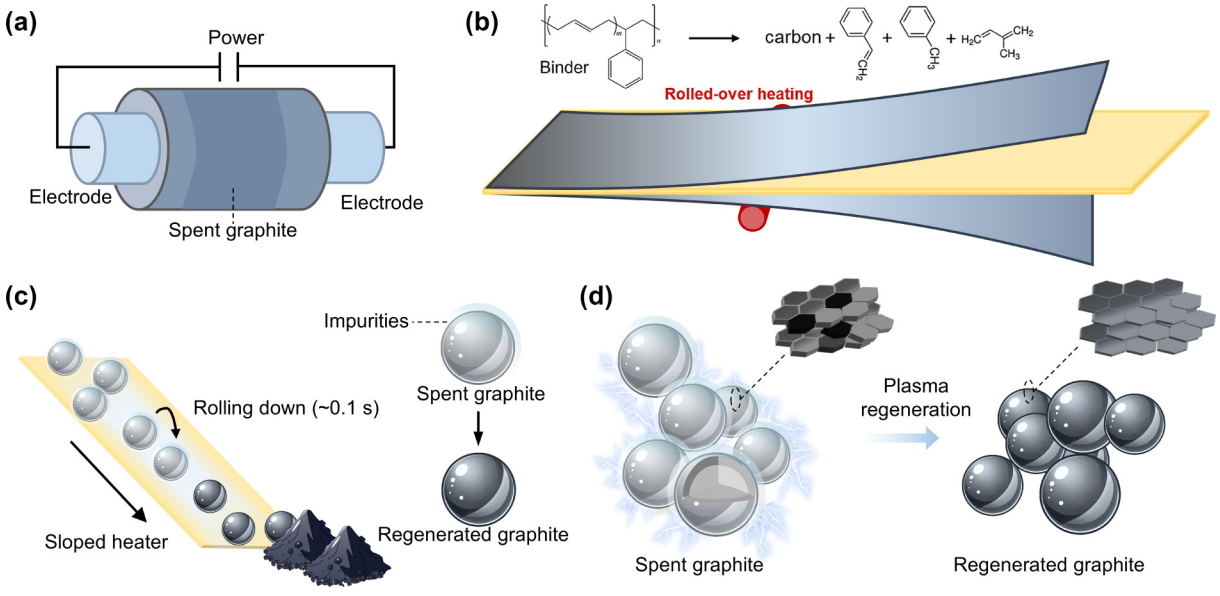

Figure 5 Recycling of graphite anode via emerging technologies. (a) Flash Joule heating for the recycling of graphite anode. Reproduced with permission from Ref. [81], © 2022 Wiley-VCH GmbH. The spent graphite is rapidly heated up within milliseconds via an electrical pulse, decomposing surface impurities and restoring graphitic order with minimal energy input. (b) Schematic of transient recycling of spent graphite from copper foils using rolled-over heating. Reproduced with permission from Ref. [82], © 2023 The Royal Society of Chemistry. (c) Schematic of the sloped carbon paper heater, where degraded graphite powder rolls down under an inert atmosphere, undergoing rapid high-temperature treatment to remove impurities such as SEI, binder, and residual electrolyte. Reproduced with permission from Ref. [80], © 2023 Li, T. Y. et al. (d) Schematic illustration of the plasma regeneration process. Reproduced with permission from Ref. [71], © 2024 Shan, M. H. et al. Plasma generated on the graphite surface enables the selective removal of surface contaminants and localized structural repair within seconds.

especially the surface residual SEI layer. Then, low cost and continuity demand graphite regeneration to evolve rapidly, posing the regeneration issues connected to surface/interface science.

5 Gap analysis between academic and industrial graphite recycling

To further analyze the difference and gap between academic and industrial recycling of graphite, the complexity of raw material, process/setup compatibility, and economic profits were compared. For scientific research, the batteries are manually disassembled into electrode sheets and other materials. The anode sheet could be immersed in water or organic solvents to remove the binder and separate the graphite on current collector. Therefore, the spent graphite material is hardly mixed with other metal impurities, besides the resolved transition metal during the battery cycling. This pure graphite powder only demonstrates some electrochemical degradation behavior, which is suitable for direct material repair. However, spent batteries suffer from simple sorting, rough crushing and sieving in the industrial recycling process. Meanwhile, the recycling companies focus on the cathode recycling and overlook the process and purity of spent graphite. The spent graphite serves as a byproduct to isolate from the mixed industrial black mass. Besides the regular degradation mechanisms, residual cathode, binder, dissolved transition metals and conductive carbon are still maintained as impurities in graphite powder, requiring complex removal processes combined with repair methods to recover its performance. Therefore, some simple strategies are difficult to satisfy the repair of industrial raw materials. It is imperative to develop precise dismantling methods in the industry to avoid electrode mixing and impurity introduction. In the repair process, simultaneous methods for removing different impurities should be further considered.

Besides the process of raw material, the process and setup compatibility also determine the feasibility of industrialization. In academic research, acid leaching and heat treatment processes are used to purify the graphite surface and repair the graphite lattice, which are compatible with industry graphite production. Although advanced Joule heating and plasma methods could reduce the repair time and energy waste in lab verification, special setups are needed to further resolve the industrial challenges. Economically, existing calculations indicate that graphite recycling delivers economic feasibility over graphite production [135]. Taking a surface repair strategy as an example [112], the regeneration process only consumes some reagents for surface reconstruction and less energy, significantly reducing the cost to \$2.48 kg^{-1}. Besides, a simple process lowers the challenge of setup compatibility. In contrast, industrial hydrometallurgy and pyrometallurgy methods require acid, alkali, a large amount of water, and extensive energy to recycle the graphite, causing a cost of ~\$11 kg^{-1}. Therefore, a targeted surface repair process could not only reduce the recycling costs but also improve the setup compatibility.

6 Conclusion and future perspective

The current industrial practice for processing spent graphite anodes predominantly relies on acid or alkali leaching, which treats graphite as a reductant or fuel source rather than aiming to restore its electrochemical functionality. This approach reflects a fundamental mismatch between recycling methods and the material requirements for high-value reuse, resulting in low-value-added products, elevated process costs, and additional environmental burdens. A mechanistic understanding of graphite degradation reveals that performance decay is primarily governed by surface-level deterioration, including the accumulation of a thick SEI, residual lithium salts, and localized lattice defects near the particle surface. In response, academic research has developed targeted regeneration strategies that selectively remove surface impurities and repair structural damage, demonstrating the technical feasibility of shifting from bulk reconstruction to precision repair. These advances underscore that the recycling approaches based on intrinsic failure modes could potentially mitigate the mismatch between the industrial graphite recycling and market demand.

Despite substantial progress in fundamental understanding and laboratory-scale techniques, the translation of these innovations into industrial practice remains limited. Bridging this gap requires a coordinated effort involving improved upstream separation to minimize impurity introduction, fundamental studies that link atomic-scale degradation mechanisms to macroscopic battery failure. Therefore, multi-scale and *in-situ* characterization technologies should be applied to the degradation research of graphite. Besides, the precise dismantling methods and technologies are important to avoid impurity introduction. Based on these, the development of advanced selective processes with simultaneous impurity removal and lattice healing should be carried out to further reduce energy and environmental footprints. Ultimately, the synergistic collaboration is critical for sustainable industry development across the entire value chain, from material suppliers and equipment manufacturers to battery producers and recycling enterprises.

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Declaration of conflicting interests

The authors declare no conflicting interests regarding the content of this article.

Data availability

All data needed to support the conclusions in the paper are presented in the manuscript and/or the Supplementary Materials. Additional data related to this paper may be requested from the corresponding author upon request.

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