Carbon Energy

WILEY

CARBON ENERGY

REVIEW OPEN ACCESS

Rethinking the Roles of Graphite and Graphene in Lithium-Ion Batteries From Environmental and Industrial Perspectives

Benjamin Robinson[1,2] | Jie Yang[1] | Rui Tan[3] | Sergey Alekseev[4] | Chee Tong John Low[1,2]

[1]Energy Innovation Centre, Warwick Manufacturing Group, University of Warwick, Coventry, UK | [2]Analytical Sciences Centre for Doctoral Training, Senate House, University of Warwick, Coventry, UK | [3]Department of Chemical Engineering, Swansea University, Swansea, UK | [4]Graphene Star Ltd, Twickenham, UK

Correspondence: Jie Yang (jie.yang.5@warwick.ac.uk) | Chee Tong John Low (C.T.J.Low@warwick.ac.uk)

Received: 19 May 2025 | **Revised:** 16 July 2025 | **Accepted:** 21 August 2025

Funding: This study was supported by European Union's Horizon Europe, UK Research and Innovation (UKRI).

Keywords: circular sustainability | graphene | graphite | green processing | net-zero

ABSTRACT

Graphite, encompassing both natural graphite and synthetic graphite, and graphene, have been extensively utilized and investigated as anode materials and additives in lithium-ion batteries (LIBs). In the pursuit of carbon neutrality, LIBs are expected to play a pivotal role in reducing CO_2 emissions by decreasing reliance on fossil fuels and enabling the integration of renewable energy sources. Owing to their technological maturity and exceptional electrochemical performance, the global production of graphite and graphene for LIBs is projected to continue expanding. Over the past decades, numerous researchers have concentrated on reducing the material and energy input whilst optimising the electrochemical performance of graphite and graphene, through novel synthesis methods and various modifications at the laboratory scale. This review provides a comprehensive examination of the manufacturing methods, environmental impact, research progress, and challenges associated with graphite and graphene in LIBs from an industrial perspective, with a particular focus on the carbon footprint of production processes. Additionally, it considers emerging challenges and future development directions of graphite and graphene, offering significant insights for ongoing and future research in the field of green LIBs.

1 | Introduction

Lithium-ion batteries (LIBs) play a critical role in contemporary technology, powering nearly all portable devices, from mobile phones to electric vehicles and, more recently, large-scale stationary energy storage systems [1–6]. The current trend of LIB production and adoption is set for continual growth and scale-up to sustain a zero-emission infrastructure. Despite the environmentally friendly aspects of LIBs, such as their use as an alternative energy storage to fossil fuels, they also present their own environmental challenges. Using life cycle assessment (LCA) data, it has been estimated that 34–77 kg CO_2 kWh^{-1} is emitted from source to product when producing LIBs [7]. This is an important factor to consider if LIBs are to continue their worldwide growth and adoption. Present LIB designs (without housing) can be broken down into four main components by weight: cathode (~41%), anode (~41%), separator (~3%), and electrolyte (~15%), as shown in Figure 1C [8]. The anode is a vital component in lithium-ion batteries (LIBs), serving as the storage site for Li ions while in the charged state. Among the various anode materials, graphite has been the predominant choice since its initial use in LIBs. Figure 2 shows the increasing trend in publications and patents relating to graphite in LIBs, which undoubtedly highlights the further growing relevance of graphite as an anode in LIBs.

https://doi.org/10.1002/cey2.70099

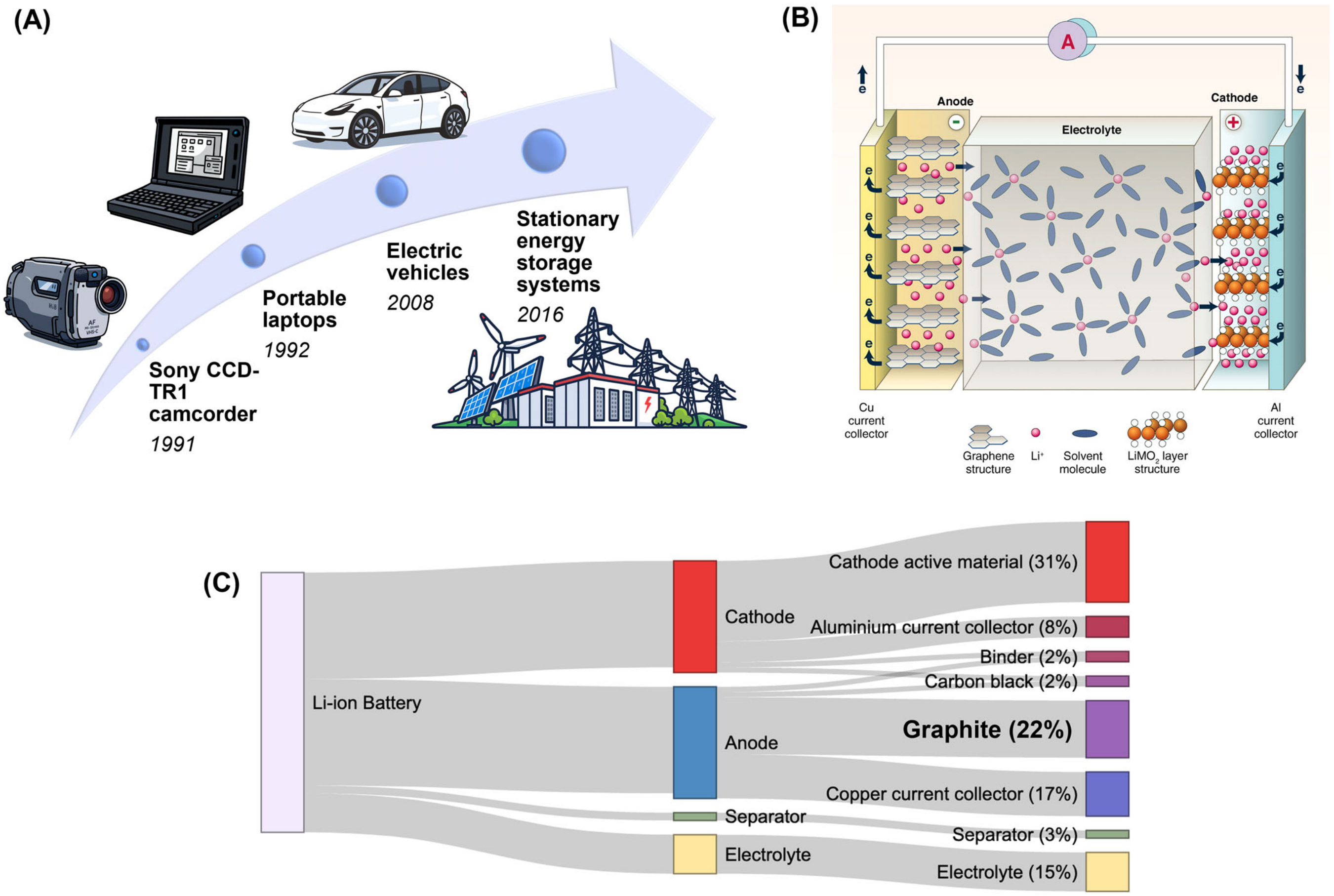

FIGURE 1 | (A) Schematic diagram showing scale-up of LIBs over the past 30+ years. (B) Schematic diagram of a lithium-ion battery. Reproduced with permission: Copyright 2011, American Association for the Advancement of Science [1]. (C) Sankey diagram breaking down the components of a modern Li-ion battery (without housing) by weight%, data taken from Yu et al. [8].

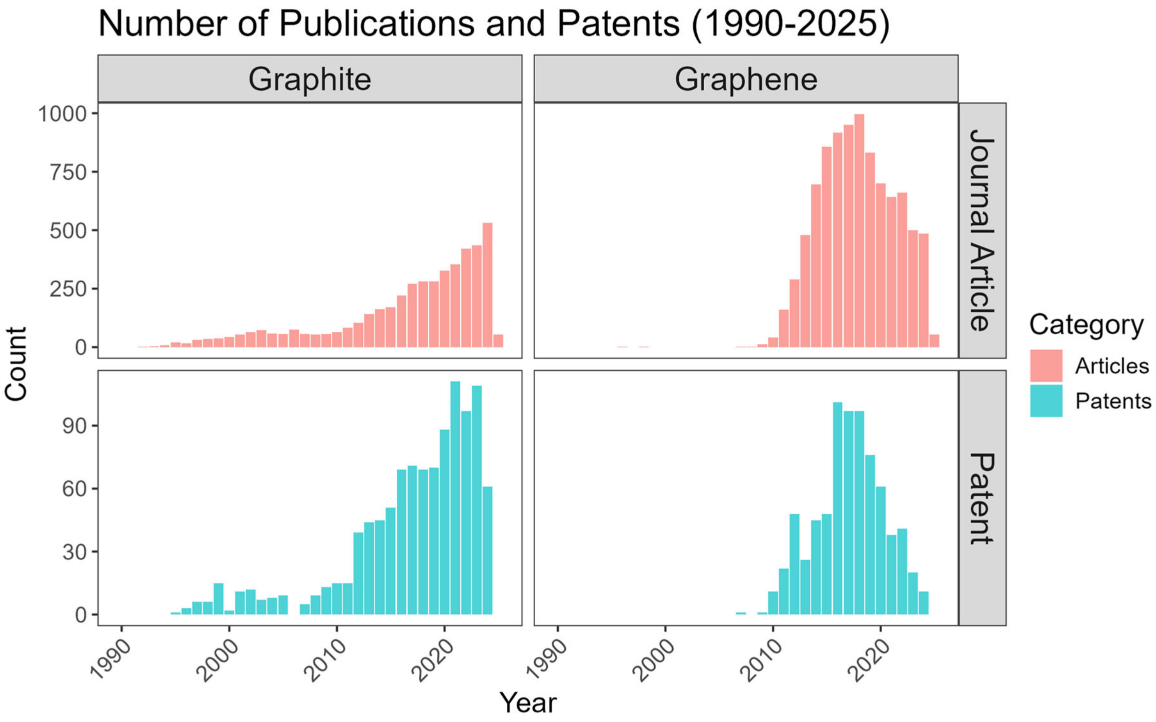

FIGURE 2 | Journal articles and patent filings relating to graphite and graphene in Li-ion batteries.

Since the commercial development of the LIB in 1991, graphite has managed to dominate the market for LIB anodes owing to its impressive theoretical gravimetric capacity (372 $\mathrm{mAh\,g^{-1}}$), high Li-ion diffusion coefficient (~$10^{-11}\ \mathrm{cm^2\,s^{-1}}$), and low delithiation potential (0.02–0.2 V vs. Li/Li^+) [9, 10]. While the chemistry of LIB cathode materials has been constantly changing from $LiCoO_2$ to $LiNi_xMn_yCo_zO_2$ to $LiFePO_4$, depending on specification demands, graphite has maintained its position as the anode material of choice, being used in nearly all LIBs sold in the market [11]. Although various types of anode materials have been widely studied [12, 13], the dominance of graphite as an anode material remains unchallenged. Graphite is expected to continue to dominate the anode market in the foreseeable future due to its technological maturity and exceptional electrochemical performance. Due to the criticality of graphite in the modern world, it has been listed as one of 17 strategic critical raw materials by the European Critical Raw Materials Act and has been defined as a mineral of high criticality by the UK government [14, 15]. It is therefore important to rethink and refine the production of graphite from both economic and environmental perspectives, thus improving the eco-friendliness of LIBs and fostering the sustainable development of society based on the principles of green chemistry.

Production of battery-grade graphite can be done by purifying and modifying natural graphite from ground deposits or by heat treatment of petroleum coke and coal tar pitch. Both methods can be energy-intensive and pollutant-heavy and part of this review aims to summarize the most recent efforts to produce battery-grade graphite via new methods. The global battery-grade graphite supply chain is complex, with a large number of companies being involved in the separate processes of natural graphite mining, petroleum coke production, coal tar production, needle coke production, synthetic graphite production, and battery-grade graphite production. Each step along the supply chain is handled by a large number of companies based in six of the seven continents of the world [16]. Battery-grade graphite production firms include: Cocan Graphite Mill Inc. (China), Shanghai Shanshan Technology Co. Ltd. (China), Shenzhen BTR New Energy Materials Inc. (China), Hitachi Chemical Co. Ltd. (Japan), JFE Chemical Corp (Japan), Asbury Graphite Mills Inc. (USA), Superior Graphite Co. (USA), and SGL Carbon SE Group (Germany). The companies listed are just a few of the 50+ companies listed as battery-grade graphite (synthetic and natural) manufacturers by Tsuji in a working paper on the global value chain of graphite in LIBs [16].

As a derivative of graphite, graphene is defined as a single layer of sp^2 carbon atoms arranged in a two-dimensional hexagonal lattice and shows promising properties for use in LIBs due to its exceptionally high surface area, electrical, and thermal conductivity [17, 18]. Graphene has found several applications in LIBs, including use as a conductive additive [19], mechanical additive [20], use in thermal management systems [21], and even proposals to use it as an active anode material [22]. However, like graphite, there is an environmental cost to producing graphene via conventional methods like chemical vapor deposition (CVD) or chemical exfoliation. The trend in publications and patents relating to graphene in LIBs can be seen in Figure 2. In both fields, the volume of published and patented work has shown a decreasing trend since 2018, after the initial boom in graphene research in 2010. Due to the peak publications reaching almost 1000 in 2018, it could be explained that, as the properties of graphene have reached maturity in academia after years of intensive research, they have therefore moved towards more industry-based research, with a focus on incorporating it into new products. Nevertheless, graphene is still used in LIBs in both academia and industry but is no longer the major focus of the most recent academic battery innovations [23].

Established companies, such as CATL, Samsung SDI, and Panasonic, have submitted patents utilizing graphene in different parts of LIBs over the past 15 years [24]. Many start-up companies have also emerged, demonstrating the capability to produce graphene at industrial scales. A meta-market analysis conducted by Schmaltz et al. found the five most mentioned graphene start-up companies in previous market reports to be: Haydale (UK), Graphenea (Spain), ACS Materials LLC (USA), XG Sciences Inc. (USA), and Nanoxplore Inc. (Canada) [25]. Other start-ups have emerged with the promise of commercial graphene-based LIBs. Granode (Sweden), founded in 2021, sells a silicon–graphene composite material with the promise of improved battery capacity, charge time, and longevity [26]. Nanotech Energy (USA) offers fast charging and non-flammable graphene-based LIBs for use in electric vehicles, home energy storage, and marine batteries [27]. These examples represent just a few of the companies that are fully utilizing graphene in LIBs, showing that the technology is already ripe for commercial applications.

For the first time, this paper aims to summarize the current problems with the manufacturing and implementation of graphite and graphene in LIBs from an economic and environmental perspective. For countries to meet their respective net-zero targets, the new technologies that will allow electrification to replace polluting methods of transport need to be sustainable. With graphite and graphene being key materials to allow the widespread adoption of electric transport, they will both require thought and consideration into their manufacturing steps to ensure they can be produced and even recycled with minimal cost, pollution, and environmental damage. Previous review papers have successfully summarized the technical challenges and merits associated with graphite and graphene performances in LIBs, but have seldom included the topic of sustainable production. For example, in 2024, Zhao et al. [28] wrote a review on the progress and challenges associated with graphite anodes in LIBs and successfully summarized the most recent research to improve the rate capability and initial coulombic efficiency of graphite anodes, but there was no mention of green processing steps to improve the industrial supply chain. Similarly, Asenbauer et al. [2] reviewed the success story of graphite as an anode material in LIBs in 2020 and contained a short section on graphite recycling but not on green processing methods; moreover, there has been significant progress made in graphite recycling worthy of review since this review was published [8, 29–31]. Likewise, recent reviews on graphene for LIB applications, such as that from Kang et al. [32], are more specifically focused on one area of graphene use, such as its synergy with silicon in anodes, rather than the environmental merits and drawbacks of its supply chain.

Graphite and graphene are also inherently linked to each other due to graphene being a single layer of graphite, meaning sustainable graphite is needed as a feedstock to produce sustainable graphene at scale. Herein, the problems and potential solutions to making graphite and graphene "green" materials for use in LIBs are summarized and discussed in detail. While existing reviews on graphite and graphene in lithium-ion batteries (LIBs) have predominantly focused on their preparation, properties, and electrochemical performance, such as capacity, cycling stability, and rate capability, this paper adopts a novel and holistic approach by integrating considerations of environmental impacts, energy consumption, supply chain dynamics, life cycle assessment, and the circular economy into the discussion. This review is particularly timely and relevant given the global emphasis on sustainable energy storage solutions. It not only summarizes the current state of knowledge regarding graphite and graphene in lithium-ion batteries but also identifies existing research gaps and future opportunities. By bridging material science with real-world challenges, this review aims to provide actionable insights for researchers, industry stakeholders, and policymakers.

2 | Current Manufacturing Methods

Graphite can be divided into two categories: natural graphite (NG), which is mined from ground deposits, and synthetic graphite (SG), which is produced by heat treatment of petroleum coke, coal-tar pitch, or oil. Scanning electron microscopy (SEM) images of particles are shown in Figure 3 [33, 34]. In short, NG has the advantage of already being preformed, with purification and shaping being the main processing methods to upgrade the natural deposits into battery-grade material. SG, on the other hand, tends to be produced to a higher purity, giving it a typically longer cycle life; there is also the advantage that it can be produced with widely available fossil fuel-based precursors, rather than the more geographically constrained natural mineral deposits. In terms of energy consumption,

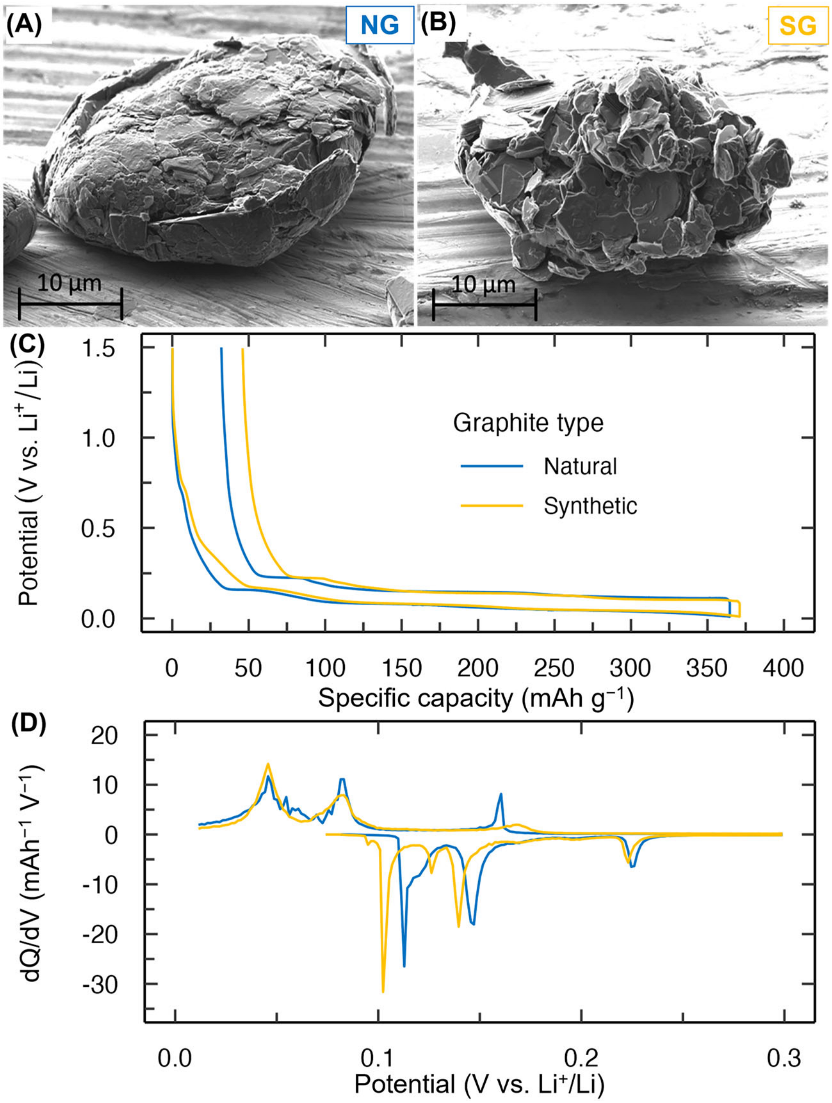

FIGURE 3 | SEM image of (A) spheroidized natural graphite particle and (B) synthetic graphite particle. (A, B) Reproduced under the terms of the CC-BY license (https://creativecommons.org/licenses/by/3.0/): Copyright 2020, The Authors, published by the Royal Society of Chemistry [2]. (C) First formation cycle of an NG and SG electrode versus Li foil with an areal capacity of 2 mAh cm^{-2} at a c-rate of C/20. (D) dQ/dV analysis of the formation profiles shown in C.

NG tends to require far less energy to produce than SG, which typically requires multiple days of processing in large Acheson furnaces at 3000°C [35, 36]. But the acid-intensive purification steps for NG tend to contribute more to local pollution as well as the emissions of more potent gases such as NO_x and SO_2 [37–39]. More specific details on the environmental differences between the production techniques are provided in the later stages of this section. Aside from the production methods, the main differences between battery-grade NG and SG are seen in the particle structure, which slightly alters its voltage profile when cycling in a LIB (shown in Figure 3). The NG has a characteristic "potato shape" from the spheroidization process, making it appear smoother in shape. NG particles also contain a higher proportion of basal planes on their surface (due to spheroidization steps) compared to SG particles, which have a higher proportion of edge planes, giving the two materials slightly different characteristics as seen in their formation profiles [2, 40]. Due to the shape difference, NG forms less of an SEI than SG owing to its lower surface area, resulting in an improved initial coulombic efficiency (ICE) in a cell, as seen in Figure 3C [41]. However, when it comes to overall performance, SG is the superior anode material, with a slightly higher gravimetric capacity and longer cycle life owing to the higher purity that can be achieved with the prolonged high-temperature pyrolysis step used in its production. The removal of impurities, such as iron, aluminum, and sulfur, is key in reducing the potential side reactions that can occur when these elements are present in the final anode material [42–44]. The two primary categories of graphite materials (i.e., NG and SG) can be further differentiated by key physicochemical characteristics, including particle size distribution, morphological features, sp^2:sp^3 carbon ratio, and purity levels. Commercial offerings demonstrate this variability; for instance, industry leader BTR markets six distinct NG grades and eight SG formulations [45], each imparting unique performance attributes to the final battery system, such as enhanced power density, improved low-temperature operation, or optimized cost efficiency. However, to maintain clarity in our subsequent discussion, we will adopt the fundamental NG/SG classification scheme rather than addressing all possible sub-varieties.

Ohta et al. [46] tested the performance of carbon-coated NG and carbon-coated SG versus Li foil in half cells using 1 M $LiClO_4$ in a 1:1 solution of ethylene carbonate and dimethyl carbonate as the electrolyte. The NG had a higher discharge capacity by ~60 mAh g^{-1} and a similar coulombic efficiency to the SG. The graphite in this experiment was not extensively cycled, and the electrolyte used is now outdated, however. A study by Glazier et al. [47] compared the performance of SG and NG in pouch cells using $LiNi_{0.5}Mn_{0.3}Co_{0.2}O_2$ (NMC 532) as the cathode material. The study showed that when using large electrolyte additive loadings (to build a more efficient SEI), the two materials showed similar coulombic efficiencies and parasitic heat flow in short-term cycling, but the NG showed poorer capacity retention over long-term cycling. When low electrolyte additive loadings were used in the cells, NG showed a lower coulombic efficiency, higher capacity fade, and more parasitic heat flow than the pouch cells containing SG. Cell stack pressure measurements showed that the NG underwent irreversible particle strain and expansion upon initial cycling of the pouch cells. This expansion was only detrimental to the performance of NG when an insufficient amount of electrolyte additive was used to form an effective SEI on the newly created surface area of the anode. NMC 622 was also studied with SG and NG as the anode materials, with NG showing a higher capacity, and lower impedance, but increased capacity fade compared to the SG [47].

A more recent study by Niu et al. [36] showed that when cycling with NMC 811 in pouch cells, NG showed a higher lithium inventory loss but also had improved electrochemical kinetics when compared to SG. The paper also showed that different SEI-forming electrolyte additives had different effects on the two different graphite materials. When using ethylene sulfate and lithium bis(oxalate) borate, more gas was formed during formation cycles with SG, but when using fluoroethylene carbonate, vinylene carbonate, and propylene sulfite, more gas was produced from the NG anode during formation cycles. The performance of NG and SG therefore varies with the cathode chemistry and the electrolyte formulations of a cell.

SG is the more popular production route for graphite across all industries, with roughly 1.8 million tonnes of SG being produced in 2021, while 1.1 million tonnes of NG were mined and processed in the same year [48, 49]. It is also claimed that currently 55% of graphite used in LIBs is SG, with the other 45% being NG, showing there is a balance between the cost and performance of the two materials, with SG being more expensive to produce but slightly better performing in the long term [49].

Table 1 shows how NG and SG compare to other emerging anode materials in LIBs in terms of commercial applications. Due to the increased purity of SG and higher production costs, it is often sold as a premium material for use in high-end electric vehicles, whereas NG is used in applications where cheaper battery feedstock materials are more viable. NG is also limited in its current production capacity due to the relatively slow process of opening new mines for production [50]. On the other hand, the feedstock materials for SG (petroleum coke and coal tar pitch) are produced in much larger quantities than NG due to their larger range of applications outside of battery technologies. This means large amounts of SG can be produced quickly to supplement any shortfalls in the total battery-grade graphite supply chain [10].

2.1 | Natural Graphite

2.1.1 | Production Overview

As reported by Engels et al. [39], there are several processes that take graphite from being a mineral in the ground to a high-value conductive anode material. This processing can be roughly broken down into five steps: mining, beneficiation, spheroidization, purification, and processing (shown in Figure 4). Graphite mineral deposits occur in three different types in the Earth's crust: amorphous, flake, and vein, with flake graphite being the optimal material for LIBs owing to its higher crystalline purity [51]. The mining of flake graphite is largely done in open-pit mine operations using drilling and blasting followed by beneficiation, which is most commonly done on site via froth flotation.

TABLE 1 | Advantages and commercial applications of NG and SG compared to other popular electrode materials used in Li-ion batteries.

	Material	Advantages	Disadvantages	Commercial applications
Anode	Natural graphite	High electrical conductivity, good capacity retention, cost-effective	Lower purity than synthetic graphite, poorer cycle life, poorer fast charge capability	Consumer electronics, electric vehicles
	Synthetic graphite	High purity, consistent performance, and better control over particle size and shape—enhancing efficiency and longevity	Higher production energy consumption and cost than natural graphite, slightly poorer coulombic efficiency than natural graphite	High-performance electric vehicles, grid storage systems
	$Li_4Ti_5O_{12}$	Exceptional cycle life, high thermal stability, fast charging capabilities	Low energy density, high cost	Electric buses, mobile medical devices
	Silicon	Higher gravimetric capacity, Earth-abundant	Excessive volume expansion, poor cycle life	Next-gen smartphones, electric vehicles
	Li metal	Highest specific capacity, lowest electrochemical potential	Dendrite formation, poor cycle life, high reactivity	High-energy-density electric vehicles, aerospace applications
Cathode	$LiCoO_2$	High energy density, good cycling performance	Most expensive cathode material, prone to thermal runaway at high state of charge	Portable electronics
	$LiNi_xMn_yCo_zO_2$	Balanced combination of high energy density, safety, and long cycle life	Complex supply chain, high cost	High-performance electric vehicles, grid energy storage
	$LiFePO_4$	Excellent thermal stability, long cycle life, and enhanced safety	Low energy density, poor conductivity	Low-performance electric vehicles, electric buses, home energy storage
	$LiMn_xFe_yPO_4$	Higher power density than $LiFePO_4$, improved thermal stability and safety, with a long cycle life	Unstable under higher C-rate charging, Poor conductivity	Power tools, electric bikes
	$LiNi_{0.5}Mn_{1.5}O_4$	High voltage and power density	Higher capacity fade, thermal instability	Power tools, electric vehicles

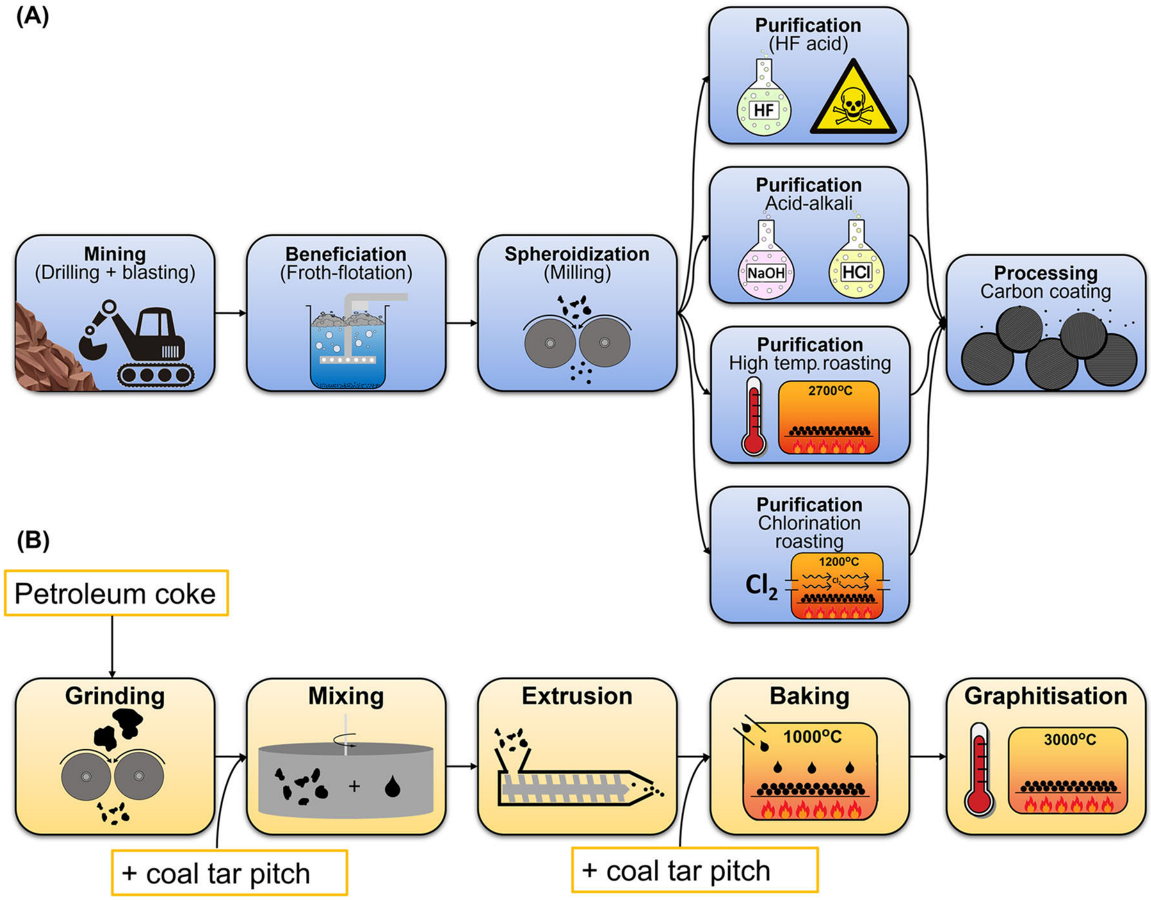

FIGURE 4 | Schematic diagram of the (A) natural graphite and (B) synthetic graphite industrial production processes currently used.

The purity of the mined graphite varies depending on the deposit location, but is usually in the range of 8%–12% graphitic carbon [52]. Beneficiation is done to remove as many of the impurities as possible by initially crushing and milling the mined deposits, followed by a multistep froth flotation process, taking advantage of graphite's hydrophobic nature to separate it from impurities.

The wet carbon concentrate is spheroidized to minimize both the particle surface area to volume ratio and the particle size distribution, which improves packing density [53, 54]. Only 40% of the graphite going into the spheroidization process comes out as spheroidized graphite, with the production scraps being classed as a by-product, which can be further processed into lower-value carbon products. Multiple classifier mills are used to micronize and spheroidize the incoming graphite in what can be a very energy-intensive step. The next step is purification, which usually aims for a 99.95% carbon purity in the graphite by using hydrofluoric acid, hydrochloric acid, and nitric acid in large quantities to leach the impurities from the graphite, which can then be removed via water scrubbing [55]. This particular method is the most popular NG purification technique used in China, with 77% of plants using this method according to Huang et al. [56] Finally, the graphite is processed to increase its conductivity by coating with high softening point pitch (derived from coal tar pitch) in a furnace at 1300°C under nitrogen gas to avoid oxidation. This coating process hardens the spheroidized graphite particles and seals their surface to minimize lithium-ion loss in the crystal structure when used in LIBs [39].

The steps listed above were from a single graphite producer and would likely be replicated with slight modifications by other graphite manufacturers. Engels et al. [39] also reported that the purification step of battery-grade natural graphite is among the most expensive and polluting of all the techniques due to the chemicals required for the process. The hydrofluoric acid method is one of four processes that are currently adopted in China to purify graphite, according to Huang et al. [56] Acid–alkali purification, high temperature purification, and chlorination roasting are also adopted industrially to purify the graphite. The percentage of Chinese manufacturers using these different techniques and their recovery rates are shown in Table 2.

2.1.2 | Environmental Impacts of Natural Graphite Production

NG production is both chemically and energetically demanding. It is important to note that over 67% of the world's NG comes from China, particularly from areas that rely on a coal-dominated energy infrastructure, causing significantly more greenhouse gas emissions [52]. LCA values of NG production from three different review papers are shown in Table 4. The most recent LCA of NG was done by Engels et al. [39] in 2022, in which data from a graphite supplier in the Heilongjiang region of China was used to predict the energy consumption and global warming potential of 1 tonne of battery-grade NG to be 10,775 kWh t^{-1} and 9.62 kg CO_2 eq, respectively.

TABLE 2 | Main NG purification methods used in China as of 2022. Recreated from Huang et al. [56].

Purification method	Capacities (kt yr^{-1})	Production (kt)	Prod.% of China	Recovery rates (%)
Hydrofluoric acid	160	48	77	68–72
Acid–alkali	30	10	16	65–69
High temperature purification	10	3	5	78–85
Chlorination roasting	2	< 1	2	73–77
Chinese total	202	62	100	65–85

TABLE 3 | Comparison of the calculated energy consumption for the different steps in battery-grade natural graphite production.

	Process stage energy consumption (%)				
Paper authors	Mining	Beneficiation	Spheroidization	Purification	Processing
Engels [39]	8.0	21.7	22.3	5.7	42.3
Gao [38]	24.6	12.5	—	21.2	41.7

TABLE 4 | Comparison of the cradle to gate (C2G) energy consumption and global warming potential of different anode grade graphite life cycle assessments.

Paper first author (Year of publication)	Production method	Estimated energy consumption (kWh t^{-1})	Estimated global warming potential (kg CO_2 eq)
Engels (2022) [39]	Natural—C2G	10,775	9.62
Gao (2018) [38]	Natural—C2G	31,244	5.32
Zhang (2018) [37]	Natural—C2G	27,906	7.75
Carrère (2024) [35]	Synthetic—C2G	24,300	42.2
Dai (2019) [57]	Synthetic—C2G	27,972	5.37
Dunn (2015) [58]	Synthetic—C2G	7,122	0.59
Notter (2010) [59]	Synthetic—C2G	20,956	2.46
Natural average		23,308	7.56
Synthetic average		20,088	12.7

A separate LCA by Gao et al. [38] was done in 2018, with the total energy use and global warming potential found to be 31,244 kWh t^{-1} and 5.32 kg CO_2 eq, respectively. A breakdown of the process stage energy consumption was also done in both papers (results shown in Table 3), and the calculated energy consumption and global warming potentials are shown in Table 4.

Both papers agree that the "processing" step of natural graphite production consumes the most energy (~42%), but there is a slight disagreement with the other steps in the process. The carbon coating processing step was found to consume the most energy due to the large energy contribution needed to heat the coal tar pitch to coat the graphite particles. Engels and Gao also analyzed different purification steps, with Engels reviewing the hydrofluoric acid method and Gao reviewing the acid–alkali method, which explains the large difference in the purification values [38, 39].

The negative effects of the use of harsh chemicals are more difficult to assess than the global warming potential of a material/process, as it can harm local environments/populations in different ways. Zhang et al. [37] found NG to have a relatively high human toxicity potential and marine toxicity potential of ~4.8 and ~2.8 (1,4-dichlorobenzene eq), respectively, which was largely attributed to the processing step of the NG manufacturing process. While these factors cause more local pollution than global pollution, they are still important factors to consider when improving via alternative NG processing methods.

2.2 | Synthetic Graphite

2.2.1 | Production Overview

SG production can also be summarized into a five-step process (shown in Figure 4). Firstly, petroleum coke (80 wt.%), produced from the residual thermal processing of crude oil, is ground and combined with coal tar pitch (20 wt.%), a steel manufacturing by-product, at 170°C, with the pitch being used as a binder for the petroleum coke [60]. Coke from coal ovens can also be used as the precursor material in this process, but petroleum coke is favored due to its higher carbon purity [58]. The resulting mixture is then formed into smaller particles using a press or extruder to maximize the density of the powder in a process similar to spheroidization [61]. The product is then baked in a furnace at up to 1000°C to further carbonize the coal tar pitch into coke. The final step of SG production, and the most energy-intensive, is graphitization in an Acheson furnace at 3000°C for 3–5 days, leaving the final SG product, which may then be later modified depending on the desired properties. The excessively high temperatures are required to both vaporize any non-carbon elements and ensure a highly ordered sp^2 carbon structure. The graphite produced from this method has a purity of over 99.9%, higher than the purity of NG (without further processing), giving it advantages when used in LIBs over the long term [2, 52, 57, 62].

2.2.2 | Environmental Impacts of Synthetic Graphite Production

Several LCAs of SG have been done in recent years (summarized in Table 4). An important trend to notice is that the global warming potentials of SG production are significantly higher in more recently written papers. Parameters that were initially left out of original LCAs were introduced into more recent ones, and local grid energy production emissions have been considered rather than national averages. For example, the work from Dai et al. expands upon the original information from Dunn et al. to include indirect emissions associated with fossil fuel production and electricity generation, while Dunn's paper only focuses on the baking and graphitization steps of SG production [57, 58]. Dai et al. calculated that 89.9 MJ kg^{-1} of energy was required to produce battery-grade SG, with 4.86 kg CO_2 eq produced from the process. This calculation was over four times higher than the calculation of Dunn et al. at 22.2 MJ kg^{-1} and was done by using China-average grid emissions data. In 2022, Surovtseva et al. [61] suggested that LCAs on SG may have been underestimating global warming potential (GWP) values by a factor of two or more. This was confirmed by a separate review of the literature and calculation on SG production by Carrère et al. [35] in 2024, which estimated the GWP to be over eightfold higher than the next most recent estimate from Dai et al. [57] Even with the assumption of all electricity being produced from hydropower, Carrère et al. [35] still predicted the GWP of SG production to be 18.1 kg CO_2 eq, still significantly larger than the work from Dai. The consumption of graphite crucibles for the graphitization process was highlighted as a key contributor to the high GWP since the crucibles must be machined from large graphite blocks and can only be reused a few times. The production and delivery of calcined petroleum coke and coal tar pitch were also significant GWP contributors outside of electricity consumption.

Aside from the energy required to produce SG, a significant number of toxic gases are produced from the graphitization process. Dunn et al. reported the emissions of NO_x, SO_x, and PM10 emissions to be 9300, 64,000, and 4100 g $tonne^{-1}$ SG, respectively, which can all be derived from impurities in the petroleum coke [57–59]. Factors such as toxic gas emissions are far harder to compare and assess on a global scale since they tend to only harm local environments instead of contributing to global warming. Local laws and regulations on toxic gases can vary greatly from region to region, meaning what is acceptable in one area may be considered extremely hazardous in another. Nevertheless, every precaution should be taken to reduce toxic emissions into local areas. This can be mitigated through the use of scrubbers, catalytic converters, or cleaning of the petroleum coke feedstock. However, all of these technologies increase the cost of the final product, and so the correct financial incentives from governments should be in place to encourage efforts to minimize local pollution.

2.3 | Comparison of Natural and Synthetic Graphite

When comparing the average results from the different LCAs, the values for each production route are quite similar. However, other papers and documents that attempt to compare both production techniques lean toward SG being more energy-intensive and environmentally damaging than NG [63, 64]. This appears more intuitive since the graphitization of SG is done at 3000°C for 3–5 days, which would be expected to consume huge amounts of energy compared to any of the other processing steps of NG. The carbon coating step of the NG also requires heating for long durations, but at a considerably lower temperature of 1300°C, so there may be a difference in database values and assumptions for the separate LCAs of battery-grade graphite. It would be most appropriate to compare the most recent LCA for each production route, as these take the previous LCAs into account and attempt to improve the accuracy of the calculations. If this is done, then the GWP of synthetic graphite far outweighs that of NG, which agrees with the papers previously cited that attempt to compare both methods.

Both battery-grade graphite production processes also suffer from poor yields. Flake graphite ore has a carbon content of 10%–15%, meaning most of the mined mass is removed in the flotation process. The spheroidization step has a yield of 40% with the low-grade “graphite fines” being sold at a lower price to other industries. Final magnetic separation and screening steps on the spheroidized graphite have a yield of 95%. The purification steps of the process can be assumed to remove impurities from the graphite flake ores that were not removed via beneficiation. Summed up, 1 kg of mined flake graphite produces 47.5 g of anode grade NG with an overall yield of 4.75% [39, 65].

For SG, the coking of crude oil gives a yield of 25% while the coal tar pitch yield from coal is 2%. The following yield of petroleum needle coke from coke via calcination is 74% and the baking/graphitization steps have a yield of 97.6%. Micronization, shaping, and coating steps may still be required; 1 kg of crude oil as the primary feedstock for SG will produce 180 g of SG with an overall yield of 18% [61]. If further modification of the SG is done, a much lower yield would be expected, although this is hard to calculate with the currently available data, and the final SG precursor would likely give higher yields in the micronization and spheroidization steps compared to NG due to the higher level of control over particle morphology before the graphitization step (Table 5).

TABLE 5 | Comparison of the production costs and material properties of battery-grade natural graphite and synthetic graphite.

Parameter	Natural graphite	Synthetic graphite
Production energy consumption (from most recently published LCA, kWh t^{-1})	10,775	24,300
Production global warming potential (from most recently published LCA, kg CO_2 eq)	9.62	42.2
Production yield (%)	4.75	18
Morphology	Rounded particles, smooth edges	Rounded particles, rough, sharp edges
Purity (carbon content %)	99.95	99.99
Typical initial coulombic efficiency (%)*	94–95	94
Typical capacity (mAh g^{-1})*	355–360	355–357
Crystalline purity (high hexagonal stacking %)	Low	High
Relative cycle life	Medium	Long

*Data taken from a reputable battery materials supplier website [45].

2.4 | Global Challenges to the Graphite Supply Chain

Due to its dominant use as an electrode material, graphite poses unique challenges in the global supply chain. As shown in Figure 5, natural graphite production is heavily dominated by China, with synthetic graphite showing a similar trend. Mozambique, Madagascar, and Brazil were the next largest producers of natural graphite in 2022, producing 170,000, 110,000, and 87,000 tonnes, respectively. With the demand for LIBs expected to soar from 700 GWh in 2022 to 4700 GWh in 2030, the demand for graphite should also match this [67]. The predicted rise in demand and dependence on graphite supply from a single country may allow political interference and resource nationalism to disrupt the global energy storage supply chain, so it is of vital importance to allow new, cheaper, and greener technologies to produce battery-grade graphite and to allow other nations to produce the materials and create a healthier and more diverse supply chain. Section 3 of this paper discusses possible solutions to help with this problem and companies that are already beginning to fix the issues.

2.5 | Graphene Manufacturing

The list of graphene production techniques is ever-expanding, with the authors of this paper counting at least 30 different techniques as of the writing of this paper. Categories of graphene production include mechanical exfoliation, chemical exfoliation, chemical synthesis, epitaxial growth, and chemical vapor deposition (CVD) [68–70]. Each technique can produce graphene with different properties, such as the number of layers, sheet size, crystal order defect density, and morphology. Due to the wide variety of uses for graphene, different applications of the material often require optimized production techniques to suit the demands of its use. The most popular methods of graphene production for applications in LIBs are liquid-phase exfoliation and the reduction of graphene oxide owing to their relatively low cost and high scalability [71]. Even when narrowing the uses of graphene to LIBs, it still shows huge versatility with applications as an anode active material, anode/cathode conductive additive, current collector support, and as a structural support and thermal distributor in thermal management systems [72–74]. As an anode active material, its high electrical conductivity and surface area can contribute to enhanced energy storage capacity and faster charge-discharge rates [22]. Figure 6 shows some of the proposed uses for graphene in LIBs.

Doping of graphene has been extensively studied as a method to enhance its electrical conductivity by modifying its electronic structure and charge carrier concentration. Through the introduction of dopants, such as nitrogen, boron, sulfur, or transition metals, the Fermi level of graphene can be shifted, altering its band structure and increasing the density of states near the Dirac point. This results in an increase in free charge carriers (electrons or holes), reducing sheet resistance and enhancing electrical conductivity. These effects collectively contribute to improved material properties, making doped graphene a potentially more suitable candidate for LIB enhancement [79]. A study by Wu et al. [80] used B-doped and N-doped graphene as an active anode material for LIBs and found it to produce a reversible capacity of $> 1040\,\mathrm{mAh\,g^{-1}}$at low charge/discharge rates. The B-doped graphene exhibited the highest ICE (55.6%), the highest power density, and the lowest cell impedance when compared to N-doped graphene and pristine graphene. Xiong et al. [81] produced N-doped graphene nanosheets, prepared by using urea to reduce graphene oxide, to be used as a cathode material for Li-ion batteries. The material exhibited a high lithiation/delithiation potential of ~3 V and a capacity of over $150\,\mathrm{mAh\,g^{-1}}$ for 1000 cycles at $1\,\mathrm{A\,g^{-1}}$. However, to be properly implemented, the material would need to be pre-lithiated before being used as a full cell cathode material, and show more capacitor-like behavior than battery-like. Wang et al. [82] produced an anode material using SnO_2 particles sandwiched between N-doped graphene layers. The material outperformed both SnO_2 and SnO_2 sandwiched in pristine graphene in lithium half cells, showing that there is an advantage to doped graphene over pristine graphene. However, the doping of graphene adds further complications to the graphene production process, with the performance benefits of doped graphene failing to outweigh the extra costs of having to dope the graphene at scale [83].

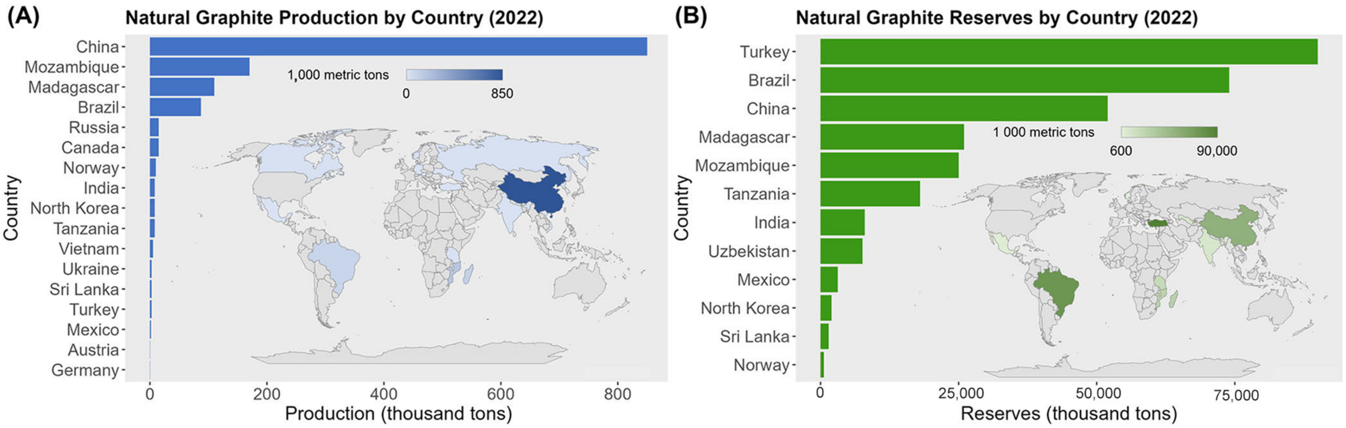

FIGURE 5 | World maps and bar charts showing the global distributions of (A) natural graphite production and (B) natural graphite reserves. Data taken from ref [66].

While the research discussed has presented doped graphene as a more favorable electrode material of choice in LIBs, the reality is that it will never be able to cycle in a full cell setup with a limited Li supply due to its poor reversibility. Papers testing the feasibility of graphene as an active anode material tend to only test the Li half cells with low mass loadings. This form of cell setup effectively provides an infinite Li-ion source and ignores the poor coulombic efficiencies that graphene suffers due to the thick SEI build-up on the large graphene surface area [22]. The only functional batteries using graphene as an active material would show a low voltage of 1 V, resulting in a poor power density and a poor cycle life due to lithium passivation [84]. Work by Chi et al. [85] found a 1:1 mix of graphene and Super P carbon black to show lower impedance when used as a conductive additive with a $LiNi_{0.8}Mn_{0.1}Co_{0.1}O_2$ (NMC 811) cathode when compared to purely using graphene or carbon black as a conductive additive in the same quantities. Graphene sheets were found to be more effective at reducing pores in the carbon-binder domain, improving the overall electrical conductivity of the cathode. However, graphene sheets were also found to wrap around the NMC particles, impeding Li-ion transport from the active material, and so a balance of graphene and carbon black gave the best cycling results [85]. Therefore, graphene alone cannot act as an effective conductive additive for LIBs. The primary uses of graphene in LIBs have been realized for many years now, which might link to the slowing down of publications in this area observed in Figure 2.

2.5.1 | Production Overview

Graphene production techniques can be broadly split into two: top-down approaches and bottom-up approaches. Top-down approaches involve the exfoliation of bulk graphite, which can be done via chemical exfoliation, edge functionalization, mechanical exfoliation, and many other techniques [86, 87]. Bottom-up approaches directly synthesize graphene sheets from precursor carbon-based molecules onto a substrate, which can later be separated to produce high-quality graphene. The two main examples of bottom-up synthesis are epitaxial growth and chemical vapor deposition [88].

As previously mentioned, liquid phase exfoliation and reduction of graphene oxide are the most popular methods of graphene preparation for use in LIBs. The chemicals used for these processes are readily available, and no specialized equipment is required, meaning labs designed for battery testing can easily accommodate the demands to produce graphene via these methods. Among current graphene synthesis methods, these two approaches emerge as particularly suitable for LIB applications due to their optimal balance between scalability, material quality, and production costs [71]. While CVD produces high-quality graphene, its superior crystalline perfection exceeds the requirements for enhancing electrode conductivity and mechanical reinforcement in LIBs, while its high cost presents a significant barrier to industrial-scale implementation [89]. Conversely, mechanical exfoliation faces challenges in maintaining consistent material quality and production scalability for battery manufacturing. These fundamental limitations render other synthesis approaches less viable for commercial LIB production compared to the two selected methods [71].

Liquid phase exfoliation usually involves the dispersion of graphite powder in an organic solvent, such as N-methyl-2-pyrrolidone (NMP), followed by an exfoliation technique, with ultrasonication and high shear mixing being the most used [90–92]. Liquid phase exfoliation comes with the disadvantages of poor particle size control and large variability in graphene sheet quality however, since the process cannot be finely tuned, with graphene flake quality varying from batch to batch. If the variability is known, though, the benefits of graphene in LIBs can still be utilized through this production method [90, 91].

The reduction of graphene oxide is done by firstly oxidizing graphite powder into graphite oxide via methods such as the modified Hummers method (shown in Figure 7B) [94, 95]. The graphite oxide should, in theory, consist of individual sheets of graphene with functionalized oxygen groups on either side of the sheet to stop them from recombining. The graphite oxide sheets are then reduced via a variety of methods, with some of the most popular being: chemical reduction with hydrazine, chemical reduction with sodium borohydride, and thermal reduction under N_2 atmosphere. The graphene produced from this technique is readily scalable and easily processed, but comes with the inevitability of oxygen impurities being left on the graphene sheets.

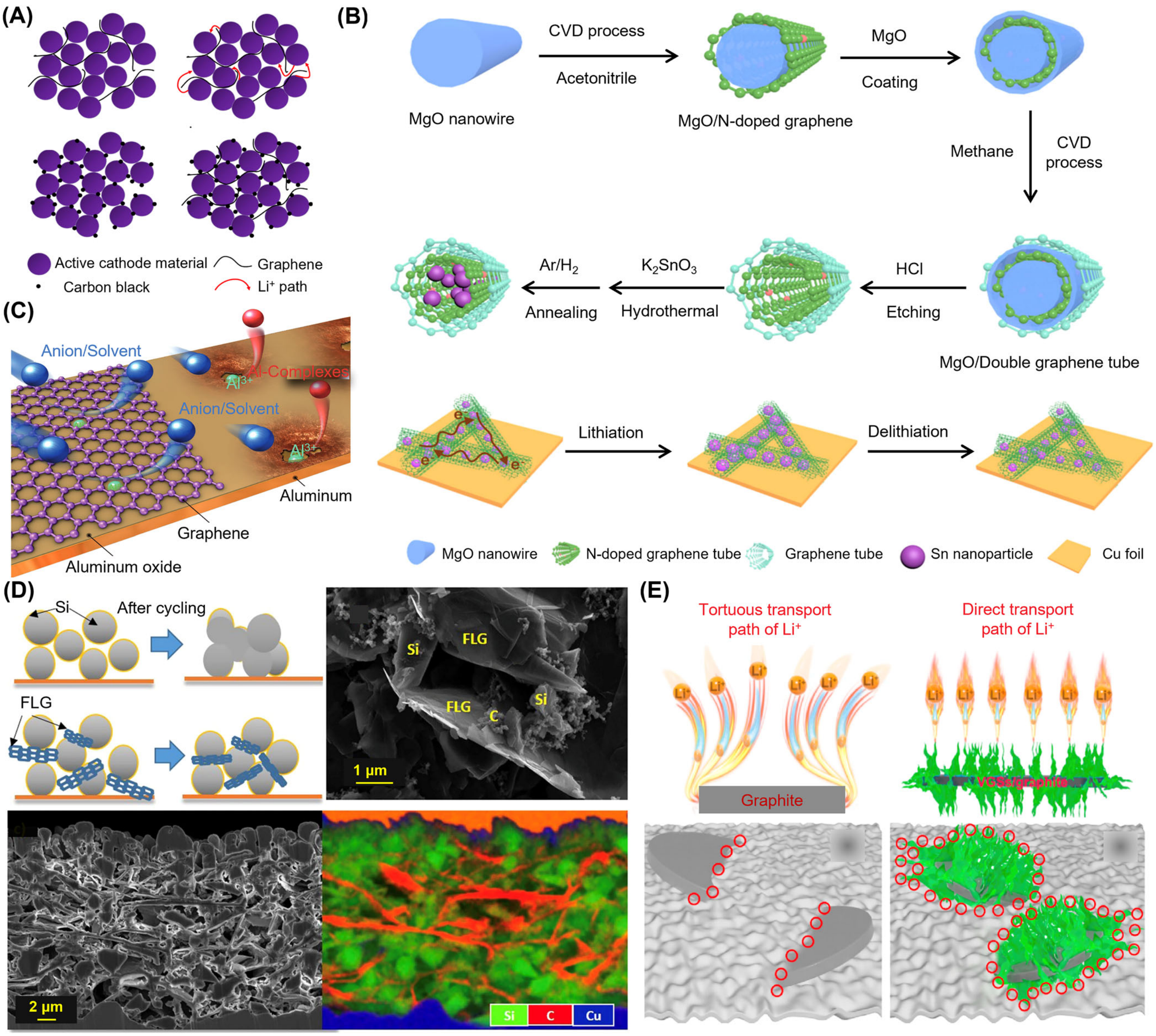

FIGURE 6 | Schematic diagrams of graphene in lithium-ion battery concepts. (A) 2D model showing desired distributions of graphene and carbon black in a cathode material structure. Reproduced with permission: Copyright 2013, Elsevier [18]. (B) Schematic diagram of the synthesis and lithiation process of Sn/double graphene tubes as a high-capacity anode material. Reproduced under the terms of the CC-BY license (https://creativecommons.org/licenses/by/4.0/): Copyright 2020, The Authors, published by Springer Nature [75]. (C) Schematic diagram of graphene armoured foil with enhanced anticorrosion properties as a current collector for LIBs. Reproduced with permission: Copyright 2017, Wiley [76]. (D) Schematic diagram, SEM and cross-sectional SEM images of Si/few-layer graphene electrodes. Reproduced with permission: Copyright 2018, Springer Nature [77]. (E) Schematic diagrams of Li ion diffusion paths in graphite and vertical graphene sheet modified graphite, along with their electrochemical contact points in red circles. Reproduced with permission: Copyright 2021, Elsevier [78].

Graphene produced via this method is commonly termed as reduced graphene oxide (rGO) to acknowledge the oxygen impurities it will contain (C/O ratio ~10) [96].

2.5.2 | Environmental Impact of Graphene Production

A LCA of graphene produced by liquid phase exfoliation of graphene oxide was done by Arvidsson et al. [97] The study used mined, unpurified NG as the precursor graphite material for both production routes, used the Hummers' method for graphite oxidation, and assumed an ultrasonication frequency of 20 kHz for 30 min. It was found that ultrasonication consumed ~470 MJ kg^{-1} graphene, and chemical reduction of graphene oxide consumed ~1,100 MJ kg^{-1} graphene. Converted to kWh t^{-1}, these values are 130,600 kWh t^{-1} and 305,600 kWh t^{-1}, respectively. Compared to graphite production, these processes demand significantly more energy to produce due to the significant energy used in ultrasonication and the high energy demand required to produce hydrazine from ammonia, used as the reducing agent.

A separate LCA of graphene production via chemical oxidation and reduction of graphene oxide was done by Lin et al. [98] with

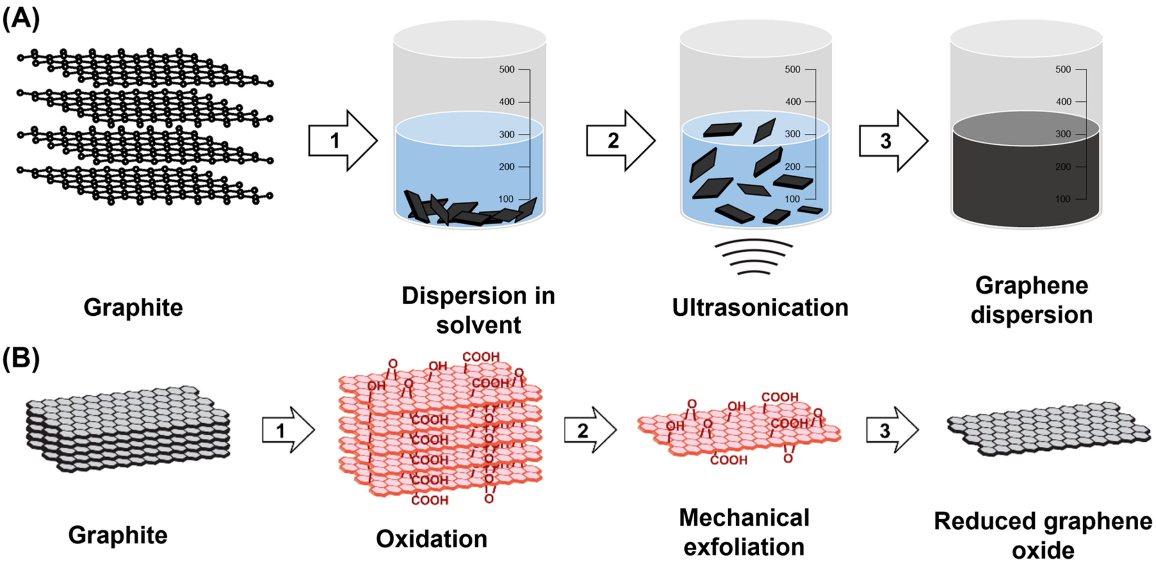

FIGURE 7 | Schematic diagrams of the production processes of graphene via (A) liquid-phase exfoliation, (B) reduction of graphene oxide. Reproduced under the terms of the CC-BY license (https://creativecommons.org/licenses/by/4.0/): Copyright 2021, The Authors, published by the Multidisciplinary Digital Publishing Institute [93].

the overall purpose of assessing the environmental impacts of the oxidative desulfurization of diesel fuel using a rGO catalyst. The paper calculated the energy usage of all the mixing, heating, and separation processes of graphene preparation to be 50,530 kWh t^{-1} rGO (182 MJ kg^{-1}) and the GWP of the whole process to be 256.5 kg CO_2 eq. The large GWP value is attributed to the large amount of hydrochloric acid that was used to wash the rGO at the end of the process, and without this, it would be reduced to ~30% of the original value [98]. The energy required to produce the precursor chemicals was not considered in the energy calculations either, with hydrazine and acids requiring significant energy to produce. The study by Lin et al. was based on the production of 1 kg of graphene in a lab-scale process. If the process were to be fully scaled up, a significant decrease in both CO_2 emissions and energy demand per kg of graphene would be expected as larger equipment would operate more economically, and solvents could be cleaned and reused between batches.

If a commercial 285 kg NMC 811//Graphite EV battery pack (70.6 kWh) with both the cathode and anode containing 1% graphene as a conductive/mechanical additive is used as an example, the total mass of graphene in the battery would be 1.66 kg (calculated using data from Winjobi et al. [99]). The 1% graphene additive would replace the 3% carbon black additive used by Winjobi et al. Using the numbers discussed earlier in this section, if the chemical oxidation/reduction of graphene oxide was used to produce the graphene, the energy demand of the battery pack would increase by 302 MJ, making graphene the most energy-demanding component of the battery pack per kg. If liquid-phase exfoliation were used, the energy demand for graphene would increase by over fivefold. For comparison, a recent LCA by Drachenfels et al. [100] calculated the total energy consumption of an NMC 811//Graphite cell from cradle to gate to be 33 kWh, translating to 8387 MJ for a 70.6 kWh battery pack. When considering that the bulk of this value is attributed to the maintenance of dry rooms and evaporation of NMP solvent in the slurry coating process, the graphene used in the LIBs would represent a high percentage of the material production energy consumption when considering that it would make up $< 1\%$ of the battery pack mass. It is clear from these brief calculations that the current methods of liquid-phase exfoliation and reduction of graphene oxide are not suitable from an ecological or environmental perspective, and newer production methods need to be designed without compromising the quality of the graphene produced.

Since graphene is poised to replace carbon black in LIB electrodes, it is important to understand the current energy and CO_2 consumption of carbon black. The C-65 carbon black used in LIBs is typically produced by the partial combustion of heavy aromatics in a furnace. Rosner et al. [101] simulated a conventional large-scale carbon black furnace production facility in the USA and found that for a plant producing 154,500 metric tonnes of carbon black, 492,000 MWh were required from the combination of natural gas and grid electricity, equivalent to 11.46 MJ kg^{-1} carbon black. The CO_2 emissions of the process were found to be 3.50 kg CO_2 eq. These values are far lower than the calculated production requirements for graphene, which were 182 and 1100 MJ kg^{-1} for oxidation/reduction of graphene oxide and liquid-phase exfoliation, respectively. If a third of the mass of graphene was required to replace the carbon black in a battery pack, the replacement of carbon black to graphene would increase the energy requirements of a 285 kg NMC 811//graphite battery pack by 173–1249 MJ and increase the CO_2 emissions by 289 kg CO_2 assuming the value provided by Lin et al. [98] It is also important to reemphasize that graphene production LCA values would likely fall significantly with production scale-up, but we still predict the values to far outweigh those of carbon black.

3 | Recent Developments

3.1 | Improved Graphite Processing Steps

As shown in Figure 4, there are many steps within the anode grade graphite supply chain, namely five steps for NG production and five steps for SG production. NG starts with mining, which has no real room for scientific innovation; instead, this process can be improved in time with more effective equipment powered by hydrogen or electricity. The beneficiation of NG improves the efficiency of the spheroidization and purification processes further down the graphite production chain. Further optimization of this step could produce an even higher-grade feedstock for the subsequent processing steps, meaning less energy input or chemicals would be required. However, the biggest areas for improvement in NG processing would be the spheroidization process (which suffers from poor yields and high energy consumption) and purification, which requires large amounts of strong acids or high-temperature roasting. However, we have found no reports of graphite purification methods that can achieve > 99% carbon wt% without the use of strong acids or temperatures < 2400°C. Green graphite purification is therefore highlighted as a key gap in the literature. The carbon coating of NG could also be another area for innovation, with carbon sources being derived from carbon-rich waste products, requiring lower heating temperatures to harden or even using processes such as flash joule heating to heat the carbon with less energy [102]. However, little innovation in greener carbon coating has been reported in the literature, with some of the most recent work still using petroleum-derived carbon sources that require high heating temperatures, so this step will not be discussed further in this section [103, 104].

As mentioned before, the current methods used for producing graphite for use in LIBs are both energy- and carbon-intensive, so new scalable methods must be developed to meet the future demand for LIBs. Often, the processing methods themselves do not produce greenhouse gas emissions, but are instead costly and energy-intensive, meaning cheaper energy is required to make the production economically viable. Since graphite production is currently dominated in regions where renewable infrastructure is lacking or where coal remains subsidized and accessible, fossil fuels are still often used to power anode-grade graphite processing facilities [105]. To make graphite production more sustainable, it must be more commercially viable in economies with a more carbon-neutral energy grid or regions that currently produce it should switch to less polluting energy production methods [106].

In recent years, several methods have been developed and commercialized to process graphite in a cleaner and more sustainable way by avoiding the use of petroleum-based chemicals and requiring lower temperatures to purify the material. Research findings are summarized below in Table 6. The technologies listed are made possible by engineering improvements to use a minimal amount of energy to either synthesize, purify, or process graphite for use in LIBs. With this technology available and set for scale-up, battery-grade graphite may be able to be produced with a considerably lower carbon footprint in the future. Further development of the technologies shown in Table 6 could also allow spent graphite from end-of-life LIBs to be processed, improving its electrochemical performance for commercial use in new LIBs and therefore building upon the circular economy.

Conventionally, mined graphite ores are upgraded with multistage froth flotation to upgrade their purity from 10%–15% to < 85% [39]. By improving the efficiency of the beneficiation stage, less chemicals will be required later in the NG purification process, improving the environmental sustainability of the NG supply chain. Ma et al. [107] proposed the use of a high-pressure grinding roller to crush flake graphite ore and a nanobubble flotation column to obtain a graphite grade of 94.82% and graphite recovery of 97.89% after three flotation stages, which were higher values than obtained with a regular 10-stage flotation process. The high-pressure grinder was able to break down the graphite ores more effectively than conventional grinding methods, and the nanobubble flotation column could effectively liberate fine graphite particles more effectively than a standard flotation column. A study by Zhang et al. [108] found that the addition of low valency cations can improve the concentration yield of graphite from froth flotation, with the addition of 0.01 M NaCl enhancing graphite concentration yield by 5.11%. The addition of cations improved the adsorption of the collecting agent (diesel) to the graphite particle surfaces, which in turn improved the flotation efficiency of the graphite. A modified thickening process was proposed by Peng et al. [109] in which a vacuum filtration step was added between the first and second flotation steps in a five-step flotation process. This increased the solid weight content of the flotation solution after the initial rough flotation step from 12% to 58.3%. The subsequent mixing of the thicker solution allowed for improved grinding efficiency, which effectively liberated interlayer impurities in the graphite ore. This process helped to upgrade the graphite purity from 91.3% to 96% after three flotation steps. Tong et al. [110] used an ultrasonication pretreatment step to increase graphite concentration yields from 89% to 95% when using an ultrasonication power of 100 W. However, this method would be hard to scale up due to the high power demand and economic challenges associated with large-scale ultrasonication.

Biber et al. successfully attempted to increase the low yield and high energy demand of natural graphite spheroidization by using the Alpine particle rounder (APR), a batch-wise operated rounding machine [111]. This new machine was able to reduce the spheroidization process energy consumption by up to 74% compared to a conventional continuous flow setup using 20–30 classifier mills (pilot scale machine shown in Figure 8C). The graphite material was tested in 0.9 Ah multilayer pouch cells with NMC-622 and showed a capacity loss of 88% after 1000 cycles, showing the new process to be mature for commercialization. The main disadvantage of this new method is that it cannot operate continuously. Further development of the machinery could be done to create a continuous spheroidization setup while minimizing the process energy consumption; solving this problem would be a key solution toward a fully green industrial graphite supply chain.

Abrego-Martinez et al. [65] used a different approach to improving the efficiency of graphite spheroidization by re-agglomerating the discarded graphite fines from the conventional spheroidization process via spray drying with citric acid and binders. When coated with coal tar pitch and heat-treated, the agglomerated graphite particles showed a comparable cycling performance to pristine commercial graphite. A slightly lower coulombic efficiency was observed due to the higher internal porosity within the re-agglomerated particles, which was susceptible to electrolyte penetration and therefore more

TABLE 6 | Commercial efforts to reduce the global warming potential of anode-grade graphite production. Companies are listed in no particular order.

Graphite manufacturer	Technology	Brief specifications	Environmental impact claims	Additionality
Green graphite technologies (NG)	Purification of natural flake graphite (GraphPure^{TM}) and renewal of secondary graphite (GraphRenew^{TM})	20,000 tonne per annum modular technology package. Graphite purity (99.95%). Tailored operating conditions for different NG sources.	No fossil fuels required. Zero carbon footprint is possible.	Only contributes to one step of the battery-grade graphite production process.
Nouveau Monde Graphite Inc. (NG)	High-grade graphite mining in Canada with spheroidization and carbon coating facilities	—	GWP of 1.23 kg CO_2 eq, very low due to cleaner energy infrastructure in Canada.	
First graphene (SG)	Conversion of petroleum into graphite and H_2 via electrochemical methods	One-step conversion performed in a hydrodynamic cavitation reactor.	No CO_2 is produced in the process.	Requires fossil fuel feedstocks.
Superior graphite (NG)	Graphite purification via resistively heated fluidized bed	Bed heated to 2400°C. Graphite purity (99.98%).	Thermal purification with comparably low emissions.	High-temperature heating will be energy-intensive.
Up catalyst (SG)	CO_2 converted into graphite via electrolysis with a molten salt synthesis reactor.	1 M tonne yr^{-1} carbon capturing capacity by 2030. 300,000 tonne yr^{-1} sustainable carbon production capacity by 2030.	Carbon negative as CO_2 is used as a feedstock.	Likely a very energy-intensive process + graphite requires further purification.
Graphene star (NG)	Processing of highly conductive natural graphite using hydrodynamic jet cavitation	0.08 kW kg^{-1} graphite power consumption.	Water-based patented method, no CO_2 produced from the process.	20 tonne per month processing facility.
CarbonScape (SG)	Produce "biographiteby processing biomass into graphite via catalytic graphitization	3 tonnes yr^{-1} production. Catalytic graphitization done at ~1500°C. 99.95% graphite purity.	Negative carbon footprint due to CO_2 sequestration in feedstock. No HF was used in the purification step.	Process has to vary according to biomass feedstock. Batch process.
EcoGraf (NG)	Developed and trademarked a HFfree™ natural graphite purification process	25,000 tonnes yr^{-1} processing capacity, works alongside the Tanzanian graphite mine, powered by hydroelectricity.	Avoids the use of HF or high-temperature treatment in purification.	Other strong acids are still required.

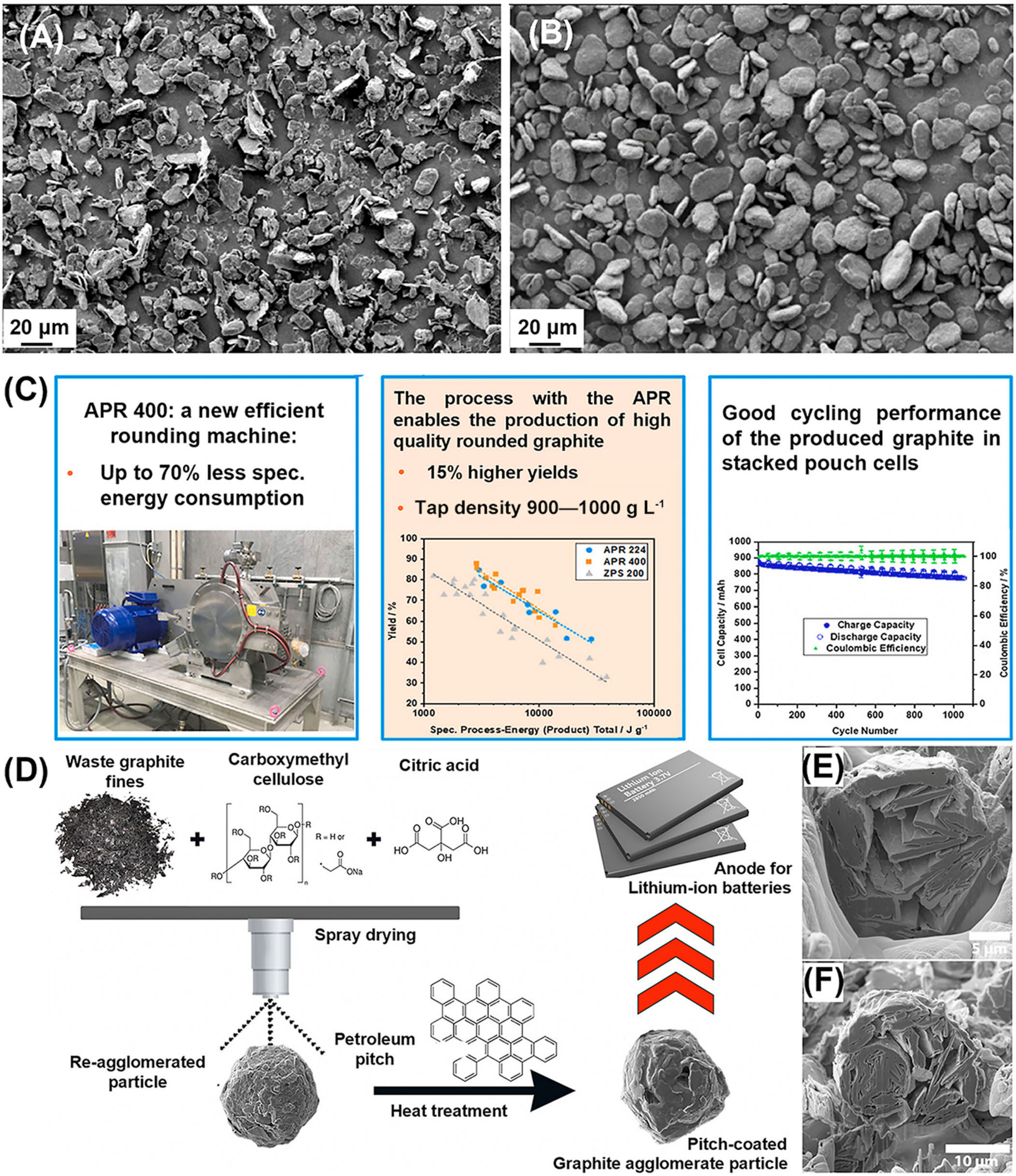

FIGURE 8 | (A) SEM image of milled natural graphite; (B) SEM image of spheroidized natural graphite from the APR machine; (C) APR 400 spheroidization machine, process energy yields, and NMC 622//spheroidized graphite pouch cell performances. (A–C) Reproduced with permission: Copyright 2023, Elsevier [111]. (D) Process of re-agglomeration of graphite fines via spray drying; (E) Cross-section of a graphite agglomerate particle; (F) Cross-section of a pitch-coated graphite agglomerate particle. (E, F) Reproduced with permission: Copyright 2023, Elsevier [65].

excessive SEI growth (shown in Figure 8F). This study helps to solve the problem of poor spheroidization yields and effectively treats the production scraps and upcycles them into higher value anode grade graphite. A step to increase the particle density could help to produce a more efficient graphite powder as well. Re-agglomeration of graphite fines can be seen as another key step to be further studied and scaled up to help produce a green graphite supply chain. The scaling up of the spray drying process may be a particularly difficult challenge owing to its relatively high cost and the need for precise control over process parameters to maintain consistency and reproducibility.

Current SG production processes start with the grinding of petroleum coke, mixing of the coke with coal tar pitch, followed by extrusion into soft carbonaceous particles of a desired size. These initial steps require simplistic equipment and require no harsh chemicals or high-temperature heating, so they have no need for scientific innovation. Instead, the baking and graphitization steps are greater areas of interest for greener processing steps, particularly for direct commercial application. Due to thermodynamic constraints, achieving graphitization of carbon typically necessitates high temperatures, making it challenging to circumvent this requirement. As shown in Table 6, commercial efforts to produce "green" SG still require heating temperatures in the range of 1500°C–2400°C. While these still seem high, they do mark an improvement to the conventional graphitization furnaces that can require 3000°C for days at a time [112]. Furthermore, waste-derived carbon feedstocks have been proposed to replace petroleum coke as a starting material, promoting the idea of a circular economy. This is discussed in the following section of this review.

3.2 | Waste-Derived Synthetic Graphite Precursors

Many processes are also being developed to produce graphite from carbon-rich waste materials, such as plastic and biomass, with low energy costs [113–115]. Similar to LIB recycling, this process utilizes the concepts of waste-to-product and upcycling, which are both valuable to the environment and the circular economy. While these processes cannot yet produce high-performing graphite in the same quantities as currently manufactured SG, they show great progress in the manufacturing of green graphite and help to build on the idea of upcycling waste materials into high-value products. More research into scalability could easily make these technologies into a commercial reality. Table 7 shows a list of different waste products that have been successfully converted into graphite and tested in Li-ion cells.

Similar to graphite recycling, the graphitic powders formed from waste material precursors tend to suffer from either a poor ICE or low gravimetric capacity. The main trends in the tested materials from Table 7, in Li half cells, were a high initial discharge capacity ($> 372\ \mathrm{mAh\ g^{-1}}$) followed by a significantly lower charge capacity. This high initial capacity is indicative of excessive side reactions on the electrode surface, which could be caused by either a high active surface area or a more amorphous carbon structure with a significant amount of sp^3 hybridized carbons or a combination of both [123]. Banek et al. [122] reported a "spheroidal" graphite derived from sawdust and algae char via a 200 W laser with a Fe catalyst. In half cells, an impressive ICE was observed (92.3%), but the cells were produced with an extremely low mass loading ($< 0.5\ \mathrm{mg\ cm^{-2}}$), likely due to the need to physically drill and skewer each particle for laser irradiation, hindering scalable production. Larger-scale cells would need to be tested to see if a high ICE and capacity retention can be achieved with more energy-dense cells. It is likely that the cells would not perform well at this scale due to the low volumetric density owing to the large pores and surface area that were observed after the biomass materials had been charred. It seems that complete graphitization of carbon-based precursors can only be achieved if the original compound structure is ordered, compact, and carbon-rich (ideally $> 90\%$), which is only the case with soft carbon and petroleum coke [124]. If a high-purity sp^2 graphite can be created, spheroidization would be the next step to improve the ICE and overall capacity of these materials in LIBs.

Other materials have also been used as precursors for SG production, although the purpose was not to create the graphite for use in LIBs. Ko et al. [115] prepared SG from polyethylene-terephthalate (PET) waste via pyrolysis and catalytic graphitization. This was done by initially carbonizing the plastic at 900°C in an inert atmosphere, followed by the use of an amorphous boron powder to graphitize the amorphous carbon in a furnace at 2400°C under a helium atmosphere. The degree of graphitization of the PET-based graphite was 80.6%, with a natural graphite sample having 89.5% and a synthetic graphite sample showing 68.9% [115]. This study holds significant promise due to the vast amounts of plastic disposed of every day, with existing processes being able to separate the plastics by type, giving a reliable feedstock material. Future research should test the anode performance of graphite derived from plastics.

3.3 | Graphite Recycling

The most economically and environmentally beneficial goal for graphite manufacturing would be to recycle it from spent LIBs, having the benefits of both reducing the amount of battery waste going to landfill or incineration, while minimizing the need to produce more battery-grade graphite. There are currently two schools of thought when it comes to the preliminary steps of battery recycling. Namely, direct recycling or shredding of battery materials. In direct recycling, cells are disassembled into their respective components to be individually processed, which represents a huge challenge when attempting to scale up the recycling process. While highly pure materials can be recovered with this method, the most promising way to safely and economically disassemble battery packs at scale would be to use robotic arms to disassemble the disregarded packs, which represents a significant challenge when different pack manufacturers and packs from crashed vehicles are considered [125]. Ideally, recycling methods will be developed that can recover all components from shredded waste LIBs (commonly termed "black mass"). The main challenges associated with recycling graphite from LIBs are: (1) Isolating the graphite from the other cathode materials and metal foils in black mass (shredding method only); (2) Removing residual binders (carboxymethyl cellulose, styrene butadiene rubber, and polyvinylidene fluoride), SEI components and excess lithium that form on the graphite anode from cycling; (3) Repairing/mitigating cracks in graphite particles and most importantly; (4) Performing all of the previously stated points in a cost effective and environmentally friendly way [62, 126]. In particular, removing the binders from black mass/recovered graphite is typically the greatest challenge, as the binders are designed to agglomerate

TABLE 7 | Summary of efforts to use waste materials as a precursor for synthetic graphite.

Carbon source	Brief description	Graphite yield (%)	Graphite purity (%)	Performance characteristics	Negatives	References
Lignin	Oxidative stabilization of lignin (heating to 250°C in air) + carbonization (heating to 2000°C under Ar).	—	—	ICE (87.9%), 126.1 mAh g^{-1} for 70 cycles at 15 mA g^{-1} (99.2% capacity retention).	Amorphous carbon in the structure results in poor capacities.	[116]
CO_2	$CO_2 + LiAlH_4$ heated to 126°C at 35 bar to produce graphite sub-microflakes.	93.7	99.988	ICE (77.5%), 320 mAh g^{-1} for 100 cycles at 100 mA g^{-1} (99% capacity retention).	Relies on large amounts of $LiAlH_4$, both expensive and highly corrosive.	[117]
Inferior coals	Inferior coals heated to 350°C under Ar + electrochemical graphitization in $CaCl_2$ at 950°C.	~52	—	~340 mAh g^{-1} at 2 C for 1000 cycles.	"Graphite" shows more similar electrochemical properties to hard carbon than graphite.	[118]
Wheat stalk	Wheat stalks heated at 150°C in water + Calcination at 800°C under N_2 + graphitization at 2600°C under Ar.	11.3	98.51	ICE (62.9%), 443.7 mAh g^{-1} at 0.1 C for 50 cycles (88.4% capacity retention).	Poor ICE, high temperatures are required to calcinate and graphitize carbon.	[119]
Used carbon cathode	Carbon cathode from Al reduction cells stirred in H_2O at 70°C + heated at 2600°C under Ar.	—	100	ICE (59.2%), 460.1 mAh g^{-1} at 0.1 C for 50 cycles (101.1% capacity retention).	Poor ICE, high temperatures are required to calcinate and graphitize carbon.	[120]
Green tea biomass	Tea leaves soaked in H_2O at 100°C + pyrolysis at 900°C under N_2 + ball milled.	—	—	ICE ($< 50\%$), 471 mAh g^{-1} at 0.1 C for 50 cycles.	Poor ICE caused by pores in the graphitic structure, significant C–O and C–H peaks observed.	[121]
Biomass (sawdust + spherical algae)	Biomass milled with Fe metal (3:1) + pyrolysis at 600°C under N_2 + diode laser irradiation at 200 W.	40%	99.95%	ICE (92.3%), 357 mAh g^{-1} at 1 C for 100 cycles.	Fe catalyst is required, which must be later separated; macroporosity limits volumetric capacity, and low mass loadings are used.	[122]

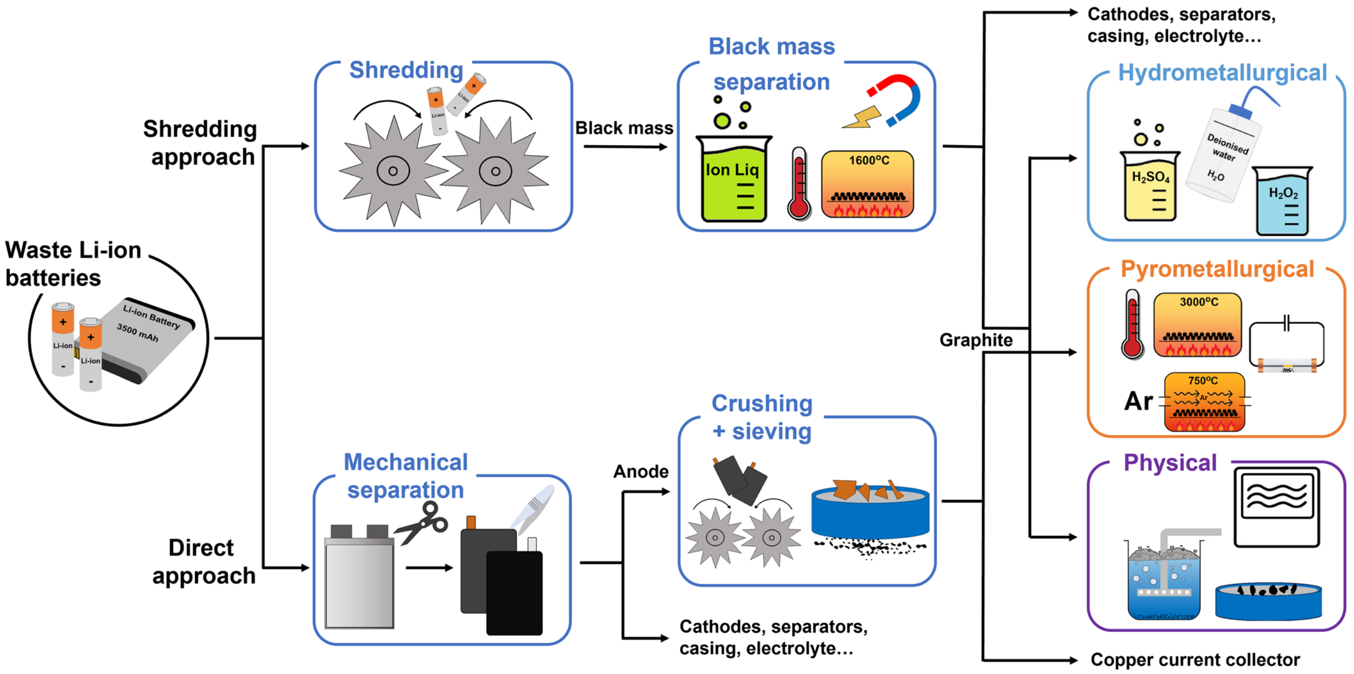

FIGURE 9 | Flowchart showing the current most popular trends in Li-ion battery and graphite recycling.

particles together, meaning different particles and current collector scraps can be difficult to separate if binders are still present in the black mass/recovered graphite. To effectively remove these, Figure 9 schematically summarizes a number of the proposed strategies to recycle graphite from LIBs.

There have been many attempts to recycle graphite recovered from direct recycling in recent years that involve using physical, pyrometallurgical, and hydrometallurgical methods [62, 127–130]. While many of these techniques have been successful at recovering graphite at a purity of up to 99.8%, none of them have proven to be cheaper or more scalable than virgin graphite production [62]. Table 8 shows some of the most recent techniques used to recycle and purify graphite that has been mechanically extracted from used LIBs.

An analysis of the results in Table 8 shows that high graphite purity can be obtained by both hydrometallurgical and pyrometallurgical recycling methods. A common issue with many recycling methods is the poor ICE of the recycled product, even when high carbon purity is achieved. This implies that many of the recycled graphite products may have had poor crystallinity, with a lower sp^2:sp^3 carbon ratio than pristine graphite. The graphite particles would have also undergone physical distortion from being cycled, creating tears within the particles [126]. Work by Dai et al. [152] showed that graphite particles expanded by 30% after 800 cycles at 1 C, with the main cause being void formation within graphite particles, giving more area for unnecessary SEI formation and parasitic side reactions. All of these factors would contribute to a low ICE [151]. It is clear that extra steps must be taken to repair the voids formed in the graphite particles for effective recycling. The most obvious solution to this would be to replicate the final steps of synthetic graphite production, where coal tar pitch is added to fill in the voids and then heated to ~3000°C to graphitize. While this would effectively repair the graphite, it would also come at a significant energy cost. Recent work by Chen et al. [153] repaired spent graphite by mixing it with pitch, from petroleum refining, in a CS_2 solution, followed by annealing at 900°C to produce a soft carbon layer. Only Li half cells were tested, and the coated graphite showed an ICE of ~83%. A similar experiment was performed by Yi et al. [154] using sucrose as the carbon source for coating, followed by sintering at 800°C. An ICE of 87% was achieved with 10 wt.% sucrose, and only half cells were tested again. Compared to the results that did not attempt an extra carbon coating step, current attempts have not been successful at improving the ICE. This is likely due to the sintering temperature not being high enough to graphitize the carbon coating, but instead creating a soft carbon layer, which improves electrode conductivity for faster c-rate charging but develops a thicker SEI layer. Both papers included a significant section on the cost analysis of the recycling processes, meaning that if a considerably higher sintering temperature were used, the process would become far more expensive and less economically viable.

Another key factor to be considered from the literature from Table 8 is the distinct lack of results using recycled graphite in full cells with a cathode. This cell setup limits the supply of Li ions in the cell, making poor coulombic efficiency a far more detrimental factor in the test. When Li metal half cells are only tested, there is a near infinite supply of Li ions in the system, meaning that Li lost in the SEI or through other side reactions can be replaced with more Li from the counter electrode. Li half cells also suffer from a poor cycle life and poor rate capability due to the significantly larger charge transfer resistance of Li metal, which dominates the cell polarization and leads to Li plating at moderate C-rates [155]. Li half cells can be improved with the addition of fluoroethylene carbonate (FEC) to the electrolyte (~15%) to provide an earlier onset of anode passivation and promote the build-up of LiF on the inner layer of the SEI [156, 157]. This Li rich inner layer allows for faster Li

TABLE 8 | Summary of the most recent graphite recycling techniques for use in Li-ion battery anodes.

Recycling technique	Classification	Graphite purity	Performance characteristics	References
Flash Joule heating at 3000 K	Pyrometallurgical	94.2%	ICE (91.7%), 350 mAh g^{-1} at 1 C 500 cycles (99% capacity retention)	[102]
Forced Li plating + water washing + graphitization at 1000°C	Hydrometallurgical	97%	—	[131]
Calcination at 1200°C + asphalt coating at 80°C + sintering at 1200°C	Pyrometallurgical	97.6%	ICE (95.5%), 394 mAh g^{-1} at 0.1 C for 110 cycles (97.8% capacity retention)	[132]
Water washing	Hydrometallurgical	~96.5%	ICE (75.9%), 345 mAh g^{-1} at 0.1 C for 100 cycles	[133]
Microwave stripping + water leaching in CO_2 atmosphere	Hydrometallurgical	< 99.3%	ICE (80.6%), 331 mAh g^{-1} at 0.5 C for 100 cycles (96.6% capacity retention)	[134]
Calcination at 1673 K for 4 h	Pyrometallurgical	99.5%	ICE (62.9%), 360.8 mAh g^{-1} at 1 C for 100 cycles	[135]
Microwave exfoliation + spray drying	Physical	—	ICE (81.6%), 409.7 mAh g^{-1} at 0.1 C for 100 cycles (106.1% capacity retention)	[136]
Leaching in 5 M H_2SO_4 and 35% w/w % H_2O_2 + filtration + sintering at 500°C	Hydro/Pyrometallurgical	—	ICE (~87%), 359.3 mAh g^{-1} at 0.2 C for 100 cycles (84.6% capacity retention)	[137]
Curing–leaching in H_2SO_4 + calcination at 1500°C	Hydro/Pyrometallurgical	99.6%	ICE (88.3%), 345 mAh g^{-1} at 0.1 C for 50 cycles (98.8% capacity retention)	[138]
Emulsification in $H_2SO_4 + H_2O_2$ + heating at 600°C + coating in pyrolytic carbon + sintering at 950°C	Hydro/Pyrometallurgical	—	ICE (92.1%), 342.9 mAh g^{-1} at 0.1 C for 50 cycles (98.8% capacity retention)	[139]
Stirring in H_2O at 1200 rpm + washing in DMC + heating at 750°C under Ar	Hydro/Pyrometallurgical	—	ICE (77.1%), 325 mAh g^{-1} at 0.1 C for 100 cycles	[30]
Leaching in 0.8 M $(NH_4)_2S_2O_8$ at 80°C	Hydrometallurgical	98.8%	ICE (80.0%), 365.3 mAh g^{-1} at 0.1 C for 100 cycles	[140]
Deep eutectic solvent treatment (choline chloride + glycerol) at 150°C	Hydrometallurgical	98.9%	ICE (88.5%), 292.9 mAh g^{-1} at 1 C for 100 cycles	[141]
Leaching in 1 M HCl at 70°C + N-doping with urea at 550°C then 800°C under Ar	Hydro/Pyrometallurgical	—	ICE (79.5%), 465.8 mAh g^{-1} at 0.1 A g^{-1} for 200 cycles	[142]
Heating at 440°C + leaching in 5 M H_2SO_4 + graphitization at 3000°C + coating with pitch at 1000°C	Hydro/Pyrometallurgical	—	ICE (90.6%), 344 mAh g^{-1} at 0.2 C for 100 cycles	[143]
Fenton reagent (Fe^{2+}/H_2O_2) assisted flotation ($LiCoO_2$ powder also recovered)	Hydrometallurgical	< 96.5%	—	[144]
Heating at 500°C + leaching in 1 M H_2SO_4 and 4% H_2O_2 at 80°C	Hydro/Pyrometallurgical	—	ICE (91.4%), 166.6 mAh g^{-1} at 1 C for 100 cycles (97.9% capacity retention)	[145]

(Continues)

TABLE 8 | (Continued)

Recycling technique	Classification	Graphite purity	Performance characteristics	References
Roasting with NH_4F at 200°C + leaching in H_2O	Hydrometallurgical	99.98%	ICE (92.1%), 135 mAh g^{-1} at 1 C for 100 cycles	[146]
Leaching in 2 M H_2SO_4 at 95°C + heating at 900°C under Ar	Hydro/Pyrometallurgical	99.96%	ICE (94.0%), 343.5 mAh g^{-1} at 0.1 C for 100 cycles (95.7% capacity retention)	[147]
Heating at 600°C + leaching in 2 M H_2SO_4 + mixing with $Co(NO_3)_2$ in alcohol (catalytic graphitization) + heating at 900°C under N_2	Hydro/Pyrometallurgical	98.1%	ICE (79.4%), 333 mAh g^{-1} at C/3 for 100 cycles	[148]
Calcination at 550°C + leaching in 1 M H_2SO_4 + microwave irradiation at 800 W for 15 s	Hydro/Pyrometallurgical + physical processing	—	ICE (84.0%), 372.9 mAh g^{-1} at 100 mA g^{-1} for 100 cycles (96% capacity retention)	[149]
Leaching in conc. H_2SO_4 + roasting at 200°C + leaching in 2 M H_2SO_4 at 90°C + microwave calcination to 800°C	Hydro/Pyrometallurgical + physical processing	—	ICE (83.4%), 354.1 mAh g^{-1} at 0.1 C for 60 cycles (98.3% capacity retention)	[150]
Washing in DEC + Washing in H_2O + plasma assisted electrolysis in 5% H_2O_2	Hydrometallurgical	98.8%	ICE (77.0%), 392 mAh g^{-1} at 0.3 A g^{-1} for 500 cycles	[151]

transport and produces a lower impedance and extended cycle life in Li half cells. Proving that the recycled graphite materials can work in a full cell setup in both coin and pouch cell formats would greatly help to prove the commercial feasibility of graphite recycling.

Common themes among the graphite recycling methodologies in Table 8 are the use of acid leaching and high temperature heating/calcination; in fact, the techniques that combined both of these methods produced the best performing recycled graphite materials. This mimics part of the manufacturing process of natural graphite, thereby potentially producing graphite with a similar purity. Future recycling processes could incorporate spheroidization and carbon coating steps to further improve the performance of the final products. Whilst there has been work on improving the efficiency of spheroidization steps on virgin graphite deposits for use in LIBs, the authors of this paper have not been able to find any graphite recycling techniques that incorporate spheroidization [111]. This area should be highlighted for future research efforts, but it is difficult to produce due to the expensive industrial equipment required.

Rey et al. [130] performed an LCA of nine different graphite recycling techniques from various papers and found the technique with the least global warming potential (GWP) to be pyrolysis and flotation, while Fenton and flotation had the highest GWP (shown in Figure 10). The main steps contributing to global warming were the use and disposal of acids and energy consumption from high-temperature heating. The method of Fenton and flotation consumed large quantities of energy from the high shear mixing required in the process and provided a poor yield of graphite, which explains its abnormally high GWP value. It would be ideal for future recycling methods to minimize the use of strong acids and high-temperature heat treatment while still producing a high-purity and crystalline graphite product. The use of catalysts may be necessary to achieve this, to reduce the high heating temperatures traditionally required to produce high-purity graphite.

3.4 | Green Graphene in the Next Generation of Li-Ion Batteries

Many patents have been submitted relating to graphene in LIBs owing to the promise of its exceptional conductivity and mechanical strength [24]. These technologies are only now starting to become a commercial reality after years of extensive research and performance optimization. The next step in the process of implementing graphene into LIBs should be implementing green graphene into LIBs. Differing production methods will inevitably create graphene with slightly different properties to graphene produced via the conventional methods discussed in Section 2. A review by Munuera et al. in 2022 concluded there were no suitable techniques to produce high-quality graphite from waste materials due to the large amounts of defects, oxidation, and impurities in the final products [158]. Table 9 summarizes recent efforts to produce green graphene without the use of conventional acids or strong reducing agents. Molten LiCl salt was used by Gürünlü et al. [160] and Kamali [163] to intercalate between the layers

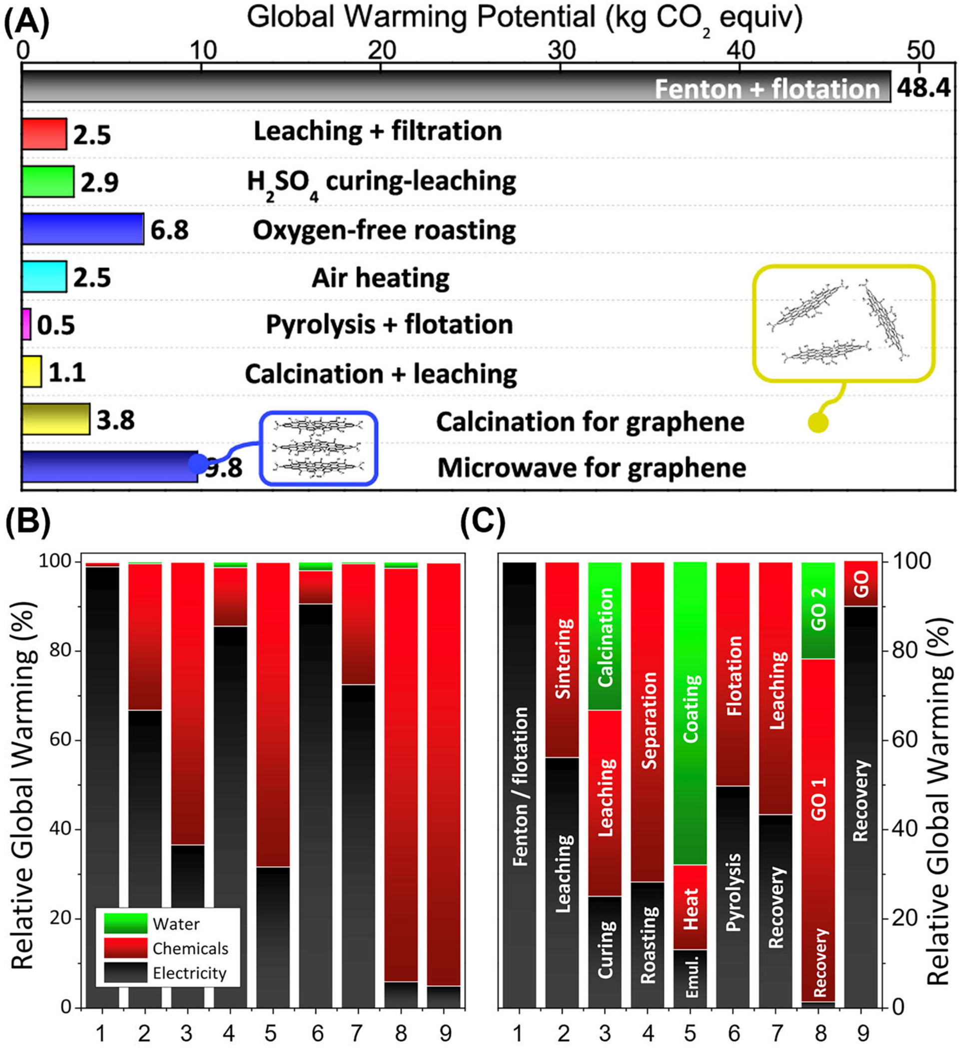

FIGURE 10 | (A) Bar chart showing global warming potential of different experimental graphite recycling techniques; (B) Relative CO_2 contribution of electricity, chemicals, and water used for each graphite recycling process from A; (C) Relative CO_2 contribution from each step during graphite recycling from A. (A–C) Reproduced under the terms of the CC-BY license (https://creativecommons.org/licenses/by/4.0/): Copyright 2021, The Authors, published by the American Chemical Society [130].

of commercial graphite electrodes and disperse graphene sheets into the ionic liquid solution. This method is capable of generating highly pure and conductive single-layer graphene while only consuming water-soluble LiCl and KCl. From a chemical consumption point of view, this is a strong example of how high-quality graphene can be produced without the need for harsh chemicals. Kamali also tested the graphene produced as a conductive/mechanical support for SnO_2 nanocrystals to produce a high-capacity anode material. Chen et al. [161] found that ultrasonication of graphite from spent LIBs has an exfoliation efficiency three to 11 times higher than standard graphite powder due to the layer expansion from the repeated (de)intercalation of Li ions in the battery's life. This also shows how spent graphite can be used to create high-value products for use in other applications. Lin et al. produced few-layer graphene (FLG) by using jet cavitation to exfoliate graphite, using significantly less energy than methods like ultrasonication. The FLG produced was also tested in Li half cells and outperformed graphite at high C-rates (5 C and 10 C); however, the FLG did exhibit a lower ICE than graphite, limiting its usage in full cells.

The examples shown in Table 9 may also show potential in other applications for LIBs. Graphene holds great promise in LIBs primarily because it serves as a superior conductive additive. Its enhanced conductivity compared to traditional carbon black opens up new opportunities for materials with previously overlooked potential as electrode materials due to their limited conductivity, granting them a fresh opportunity to become active electrode materials. Figure 11 shows a small selection of new electrode materials that have been made viable in LIBs by graphene.

Another key limitation of many electroactive materials is the considerable volume expansion that occurs after Li-ion intercalation. A high gravimetric capacity often comes with a high-volume expansion due to the increased number of Li-ions in the material at full capacity. Si undergoes an expansion of up to 300% when fully lithiated, and sulfur expands by up to 80% [167, 168]. This increased expansion leads to both safety issues within a battery and significant capacity degradation upon initial cycles of the cell due to damage and reforming of the SEI. Small quantities of graphene can be used to cushion volume

TABLE 9 | Summary of green methods to produce high quality graphene.

Method description	Positives	Negatives	Graphene properties	References
Exfoliation of graphite in water with tannic acid as dispersant and interfacial regulator	Tannic acid can be found in most plants	Sonication for 1 h required. Tannic acid is still attached to graphene, not pure.	Electric conductivity of 48,800 S m^{-1}. Graphite exfoliation efficiency of 2.5%.	[159]
Exfoliation of graphite in molten LiCl–KCl	Only consumables are graphite and electricity	High temperatures are required to melt salt (600°C). Oxygen-free atmosphere is required.	Electric conductivity 1219 S m^{-1}. Carbon content of 86%.	[160]
Exfoliation of spent graphite from LIBs via horn sonication	LIB cycling increases interlayer distance in graphite, allowing improved exfoliation efficiency	Graphene still requires annealing at 500°C. Sonication required.	Electric conductivity of 9100 S m^{-1}. Graphene mass yield of ~40%.	[161]
Jet cavitation liquid exfoliation to produce few-layer graphene	Deionized water is the only chemical used	Single-layer graphene was not produced.	Cycled in Li half cells as an active anode material. ICE (90%), 200 mAh g^{-1} capacity at 10 C.	[162]
Exfoliation of industrial graphite electrodes under cathodic polarization in molten LiCl	Only consumables are graphite, H_2, and electricity	High temperatures are required to melt salt (800°C). Oxygen-free atmosphere required.	Electric conductivity of 580,000 S m^{-1}. SnO_2 nanocrystals anchored to graphene and displayed Li capacities of 1016 mAh g^{-1} after 100 cycles at 1 C.	[163]

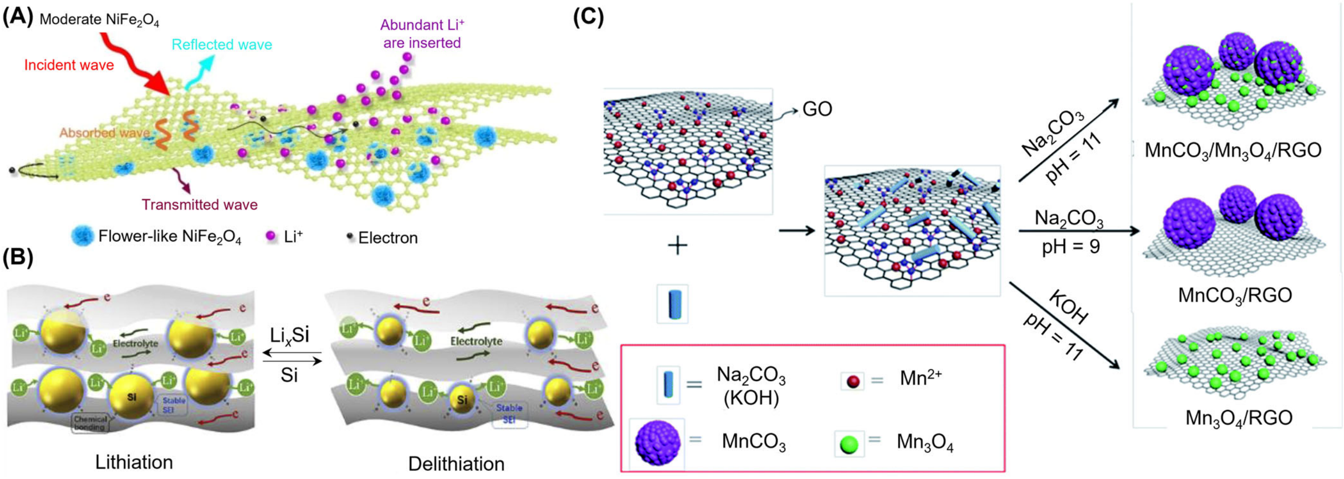

FIGURE 11 | Schematic diagrams of graphene enabling the application of new electrode materials in LIBs. (A) Lithium storage mechanism of $NiFe_2O_4$/graphene composite. Reproduced with permission: Copyright 2022, Elsevier [164]. (B) Mechanism of Si nanoparticle/citric acid/graphene oxide composite electrode during lithiation/delithiation. Reproduced with permission: Copyright 2020, Elsevier [165]. (C) Structural evolution of multicomponent Mn-based composites. Reproduced with permission: Copyright 2017, Royal Society of Chemistry [166].

expansion of these materials by creating a stable and highly porous electrode microstructure that can facilitate significant volume expansion [32]. Chen et al. [169] successfully produced a mesoporous 3D graphene structure with Si nanoparticles deposited on it, in an effort to reduce the pulverization of the Si active material, and achieved a capacity of 1200 mAh g^{-1}. Papandrea et al. [170] used a similar 3D graphene structure to mechanically and electro-conductively support a Li-sulfur cathode to achieve a gravimetric capacity of 969 mAh g^{-1}, a higher gravimetric cathode capacity than any previously reported sulfur cathode.

Due to the high commercial relevance of both graphene and LIBs, some of the most promising graphene/LIB efforts have been commercialized by both start-up companies and established technology companies, such as Samsung and Huaweii [171, 172]. Table 10 lists existing companies that have commercialized or are in the process of commercializing graphene into LIB technologies. Samsung, Lyten, and Levidian all claim to produce their graphene via CVD of methane onto a metal substrate. This allows the companies to claim that they are using greenhouse gases as a feedstock to produce both graphene for more highly performing batteries and H_2 gas for use as a green fuel. However, the energy input for CVD is significantly high, necessitating the use of green energy sources to ensure the process is fully sustainable.

4 | Remaining Challenges

4.1 | Integration With Existing Manufacturing Processes

As of 2022, the annual demand for battery-grade graphite was approximately 130–210k tonnes, with the demand expected to be more than three times higher by 2030 [176]. Due to current industrial methods being well established and primarily based in China, substantial investment is required elsewhere in the world to diversify the supply chain and keep up with the projected demand. Setting up new production facilities for both SG and NG will be a time-consuming and capital-heavy task, while turning a profit may be even harder with energy prices being considerably more expensive in Western countries compared to China [177]. For a population to adopt electric vehicles, the cost of the materials that go into the batteries needs to be kept as low as possible so that consumers can afford to buy them.

In the previous section of this paper, multiple technologies and processes that have been developed and utilized by separate research groups and companies have been discussed. Combining these various innovations together would massively help to produce a healthy supply chain for green graphite production. Ideally, NG would be sourced from flake graphite deposits in countries such as Turkey, Mozambique, and Canada, with the following processing steps being used as close to the graphite mines as possible to minimize transport costs and emissions. Next, beneficiation via froth flotation should be used to crudely remove impurities, followed by the GraphPure^{TM} purification process utilized by Green Graphite Technologies to reach a purity of 99.95%. The spheroidization process proposed by Biber et al. [111] could then be used to spheroidize the graphite particles with a far higher yield and lower energy input than current spheroidization methods. The waste graphite fines from this step could be converted into suitable graphite particles with the re-agglomeration method proposed by Abrego-Martinez et al. [65] to further increase yields. Finally, the carbon coating step can be done on the final graphite particles to produce a green NG product ready to be utilized in LIBs for energy storage. This proposed method is idealistic, however, since the various companies associated with these different technologies are from different parts of the globe, with many of the processes still being in their pilot phase. Opening new open-pit mines is also a lengthy process with environmental and regulatory approvals required, as well as local infrastructure developments and sufficient financial investment [50]. It is also important to note that the battery-grade graphite supply from China should not slow down, but instead aim for lower GWP and energy

TABLE 10 | Summary of companies claiming to use graphene to improve the performance of LIBs.

Company	Technology	Key metrics	Comments
Nanotech energy	Graphene enhanced cathode + anode	Flame-resistant, "ultra-fast" charging	Graphene produced from reduced graphene oxide
Granode	Graphene cages containing Si nanoparticles	28% higher energy density, 85% lower carbon footprint	—
Samsung	Graphene balls, graphene-silica assembly	716 mAh g^{-1} as anode active material, 78.6% capacity retention after 500 cycles at 5 C and 60°C as cathode conductive additive [171]	Synthesized via chemical vapor deposition of CH_4 onto SiO_2 nanoparticles
Huawei	Graphene-grafted polymer conductive additives for electrode plates	Capable of cycling at 60°C, full charge at 3 C (3 Ah battery), and double lifetime (compared to the previous model) [172]	Originally released in 2016, still used in current Huawei phones
Lyten	Sulfur cathode with 3D graphene composite	Mitigation of polysulfide shuttle effect, 1640 Wh $kg^{-1}{}_{cathode}$, higher charge/discharge rates enabled, uses Li metal composite anode [173]	Graphene produced from greenhouse gases, all cylindrical cell parts sourced from the USA, Norway, and India. C2G emissions 25 kg CO_2 eq kWh^{-1}
Appear Inc.	Flexible graphene-based LIBs for portable devices	Full charge at 3 C	—
Targray Group	Graphene anode powder	55% graphene by area, 0.06–5 μm particle size, offered in suspension or powdered form	Produced via a scalable method from ultra-pure graphite (99.995%) without the use of acids or oxidants
Levidian	Graphene supplier, used as a conductive additive in anodes	—	Graphene produced from the splitting of CH_4 [174]
Graphenano Energy	Graphene fiber ink current collectors	Improved energy density, cell potential can be increased by 0.5 V due to the avoidance of high localized conductivity [175]	—

consumption values to maintain the essential supplies that LIB manufacturers rely on across the world.

Alternatively, spent graphite from waste LIBs could be used as feedstock materials. However, isolating the graphite from the spent LIBs at an industrial scale poses significant challenges. The most promising method to do this is via froth flotation (FF) of LIB black mass to take advantage of the difference in hydrophobicity between graphite and cathode materials. FF is a well-established and widely utilized technique within the mining industry. Consequently, significant research has already been conducted to scale up and optimize the process, ensuring its efficiency and applicability on an industrial scale. Much research on the use of FF to liberate graphite from black mass has been done by Vanderbruggen et al. [178–181], where it has been found that adding an attrition pretreatment step can break agglomerates and increase the particle surface areas available to the FF treatment, increasing the flotation efficiency of graphite to 85% (shown in Figure 12). Furthermore, work by Olutogun et al. [29] used graphite recovered from this process and then purified it via mixing with caustic soda and heating to 500°C, followed by mixing with dilute sulfuric acid and washing with water to produce a final carbon purity of 99.2%. The recovered graphite was found to have a remarkably similar structure to pristine graphite and demonstrated a reversible capacity of > 350 mAh g^{-1} in half cells and an ICE of 86.6% in full cells with an NMC_{532} cathode. To the best of our knowledge, this is the only current example in the literature of graphite recovered from black mass being successfully cycled in a full cell format.

The work from Olutogun et al. [29] is a promising example of a true full-cycle reuse of graphite in LIBs and holds great hope for future work. Future work could include the testing of these materials in the more industrially relevant pouch cell format to fully prove the commercial feasibility of the process. Additional work could also be done to test different pretreatment methods on black mass before froth flotation, as this seems to be a key area for improvement to enhance the flotation efficiency of recovered graphite. Many of the methods proposed in Table 8 could be directly applied to black mass before FF treatment, since they attempt to remove the binders from the graphite, which would help to liberate graphite, cathode, and current

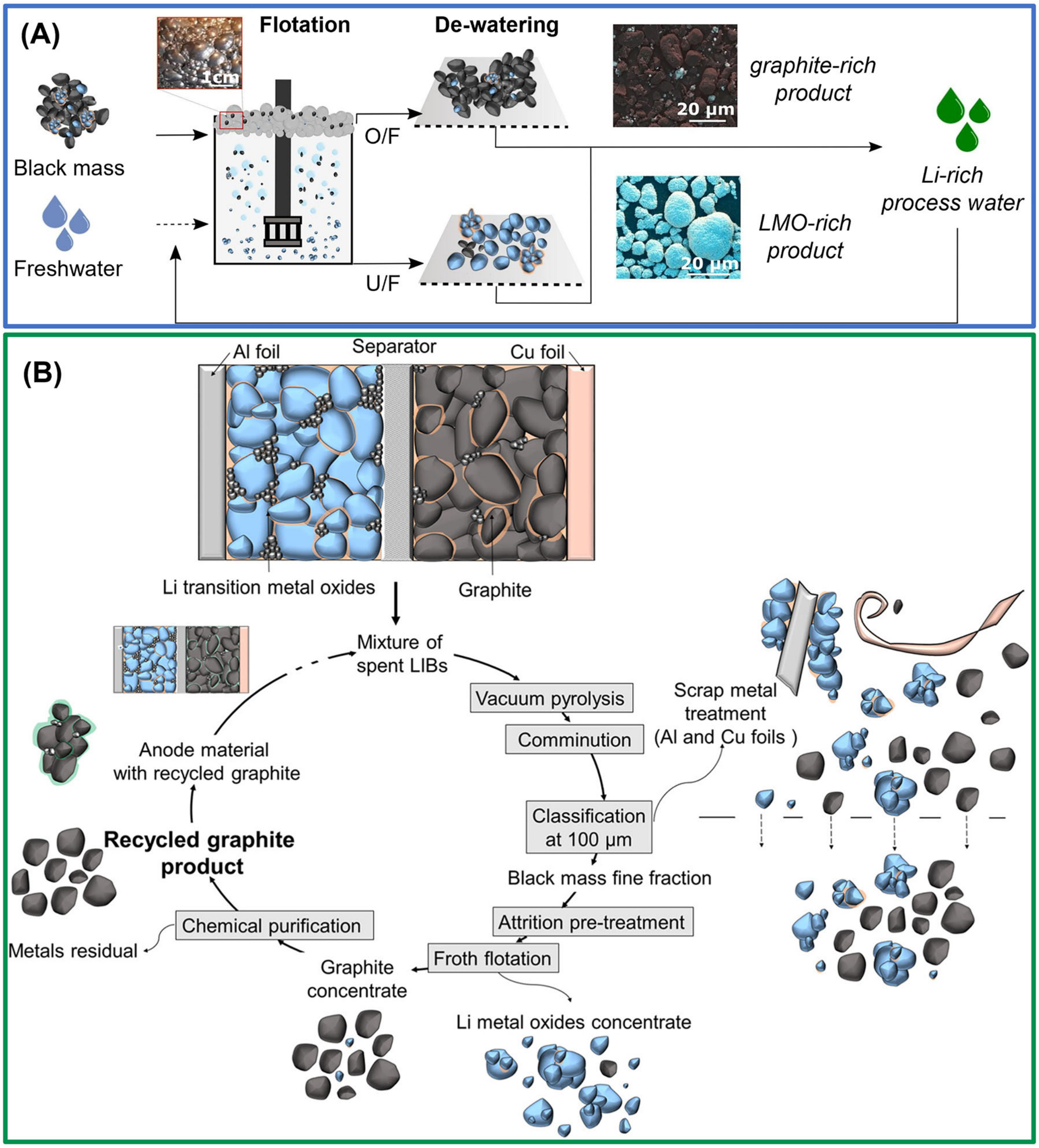

FIGURE 12 | Schematic illustration of (A) the froth flotation process used to recycle graphite from black mass. Reproduced with permission: Copyright 2022, Elsevier [179]. (B) Closed loop cycle proposed for recycling graphite using froth flotation as a key step to separate graphite from black mass. Reproduced under the terms of the CC-BY license (https://creativecommons.org/licenses/by/4.0/): Copyright 2024, The Authors, published by John Wiley and Sons [29].

collector particles from each other, allowing for more efficient separation. The pretreatment steps of vacuum pyrolysis, comminution, classification, and attrition proposed by Olutogun et al. [29] are clearly strong methods, but more energy-efficient and higher-yielding methods could also be explored.

4.2 | Graphite Disposal Methods

When compared to the other components of LIBs, graphite presents a relatively low ecological risk due to its relative inertness and low reactivity. Due to the toxic chemistry and high value of many of the components in LIBs, they must be disposed of in a safe and effective manner to avoid these components ending up in landfill sites and leaking into nearby ecosystems [182]. Currently used recycling techniques that aim to recover cathode materials and current collectors of LIBs can be divided into pyrometallurgical, hydrometallurgical, and physical processes [31, 127]. Pyrometallurgy involves heating the waste components of LIBs to “burn off” the lighter and less valuable materials, such as the separator, polymer binders, and carbon-based components; this is an effective technique for targeting the extraction of the critical metals in the battery, but when done in the presence of oxygen the graphite in the battery will combust according to equations (1) and (2).

$$C_6 + 6O_{2(g)} \rightarrow 6CO_{2(g)} \quad (1)$$

$$C_6 + 3O_2 \rightarrow 6CO_{(g)} \quad (2)$$

Assuming complete combustion of the graphite in this process, 1 tonne of graphite burnt will produce 3.67 tonnes CO_2 eq with a plentiful supply of oxygen. The energy from this process could be used to effectively downcycle the waste graphite material into usable energy, but the wasted graphite would still produce significant greenhouse emissions as well as other chemical pollutants from the SEI, so it would not be feasible. On a larger scale, this process would contribute a significant amount toward greenhouse gas emissions, partially mitigating the environmental benefits of LIBs. Methods to recycle the graphite from LIBs have already been discussed to prevent its incineration or burial.

Spent graphite from LIBs has also proven to be an effective feedstock material for graphene exfoliation, owing to the reduction of the interlamination force from repeated Li-ion (de-)intercalation [183]. Table 11 shows a summary of different methods used to produce graphene from spent graphite. Zhang et al. [186] found that H_2SO_4 and $KMnO_4$ consumption were 40% and 28.6% lower when using the simplified Hummers method to produce graphene oxide from spent graphite compared to pristine graphite. This helps to further highlight the commercial value of graphite from spent LIBs. However, most of the processes discussed in Table 11 lack the scalability of industrial graphite production due to the fact that graphite had to be manually extracted from the spent LIBs as opposed to being extracted from black mass (only done by Xie et al. [189]), which can be rapidly produced from all LIB formats. Ideally, greener processes such as froth flotation could be used to separate the graphite from black mass components instead of acid leaching, used by Xie et al. [180, 189]. Nevertheless, using black mass as a feedstock material holds the most value for scalable green graphene production. Converting the recovered graphite into high-value graphene at scale would solve the problems of waste graphite disposal, the need to purchase pristine graphite, and reduce chemical/energy costs. This all feeds into the concept of upcycling the waste graphite into a higher value material, resulting in a far higher economic benefit, with the resultant graphene possibly being used to improve the performance of future LIBs [190].

4.3 | Graphene Recycling and Disposal

With the most promising uses of graphene in LIBs being as additives and its chemical similarity to graphite, it is unlikely that graphene will be able to be effectively recovered from waste LIBs. Given that it is classed as a high aspect ratio nanomaterial (HARN), there may be challenges in the safe disposal of the material from waste LIBs, although there are other chemicals in conventional LIBs that represent a significantly higher hazard to the environment, such as electrolyte solvents or Ni-rich layered oxides [191]. The most likely scenario for graphene disposal from LIBs will be the same as carbon black, incineration during pyrolytic recycling steps. Due to the weight of graphene in future LIB packs likely being $< 1\%$, there would be little financial or environmental advantage of attempting to recover the material, with recycling processes likely outweighing the economic and environmental cost of burning off the waste graphene and synthesizing new material. If LIB recycling processes avoid high-temperature pyrolysis, then perhaps the graphene could remain with the electrode materials during recycling so that when the materials are remanufactured into electrodes, they will come premixed with graphene as an additive.

4.4 | Cost and Economic Viability

Without government intervention, producing greener graphite and graphene for the LIB supply chain needs to be either cheaper or identical in price to current manufacturing processes. Battery-grade graphite costs approximately \$8000–\$13,000 per tonne, meaning newer recycling/processing/production processes should aim to match or undercut this mark to make the products economically viable [192]. A review by Liu et al. [193] concluded that graphite recycling was not commercially feasible yet since there was no existing method that was able to balance the relationship between pollution and cost. Reducing or eliminating the use of strong acids and reducing the complexity of existing processes were highlighted as the key to commercial graphite recycling success. As discussed earlier, both spheroidization and carbon coating need to be done to recycled graphite powders to improve upon or match the performance of pristine commercial graphite [46, 53]. Other green processing techniques that can be applied to virgin graphite materials show a more rapid pathway to commercial success, such as the more efficient spheroidization step shown by Biber et al. [111] or the HF-free purification method developed by EcoGraf. Furthermore, a more simplistic way to reduce the GWP of virgin graphite is to employ similar mining and processing methods in areas that rely on a more renewable energy supply, such as what is being done by Nouveau Monde Graphite Inc. in Canada, which boasted a GWP of 1.23 kg CO_2 eq, 11 times lower than a similar review of graphite from China [194]. The production processes for the products are largely the same, but run off separate energy grids.

5 | Conclusions and Perspectives

This review has systematically examined the key environmental challenges facing the global supply chains for battery-grade graphite and graphene and explored potential innovative solutions emerging from both academic and industrial research. As lithium-ion batteries (LIBs) play an increasingly central role in achieving net-zero goals, the projected exponential growth in demand for high-quality graphite and graphene necessitates urgent attention to their production sustainability by minimizing carbon footprint. Although current production processes contribute modestly to global CO_2 emissions, anticipated scale-up scenarios underscore the importance of pre-emptively addressing their environmental impact. In response, numerous startups and research groups have proposed solutions aimed at improving the sustainability and efficiency of the graphite and graphene supply chains. These include using renewable energy-powered mining equipment and improving graphite processing methods to increase yields while reducing energy consumption.

TABLE 11 | Summary of methods of producing graphene from spent graphite from waste LIBs.

Method description	Positives	Negatives	Graphene properties	References
Charged LIBs were manually dismantled, anodes shredded, sieved added to deionized water, ultrasonicated in NMP, and Li_2CO_3 removed via precipitation	Successfully extracts Li from LIBs as well as graphene, and works with medium-sized quantities of waste material	Danger in disassembling LIBs when fully charged, NMP is both expensive and toxic	1–2-layer graphene produced, breakeven price of graphene \$540 tonne^{-1}	[184]
Manual disassembly of graphite from LIBs, modified Hummers method used to simultaneously remove impurities from the graphitic structure and produce graphene oxide	Graphite simultaneously ”cleaned” and converted to graphene oxide	Conc. H_2SO_4 and H_2O_2 are required for Hummers' method, expensive and corrosive	Graphene oxide (63–67 C/at%), with specific surface areas 100–200 times greater than waste graphite materials	[185]
Manual disassembly of graphite from LIBs, Calcination at 600°C for 1 h, simplified Hummers method, ultrasonication, reduction in N_2H_4	H_2SO_4 and $KMnO_4$ consumption is 40% and 28.6% lower compared to graphene preparation from clean graphite	Conc. H_2SO_4 and H_2O_2 are required for Hummers' method, and N_2H_4 for reduction, expensive and corrosive	Optimum I_D/I_G ratio of 2.56	[186]
Manual disassembly of graphite from LIBs, modified Hummers method, reduction in molten NaOH–KOH eutectic at 220°C for 10 h	Reduction of graphene oxide does not require N_2H_4	Conc. H_2SO_4 and H_2O_2 are required for Hummers' method, an expensive and corrosive process that has many steps and requires heating for extensive periods	Colloidal dispersion in ethanol and water remains well dispersed after 4 weeks	[187]
Manual disassembly of graphite from LIBs, mechanical milling, Freeze-thaw processing in deionized water, and ultrasonic-assisted circulation	Graphene flakes from spent graphite showed better morphology than graphene flakes prepared from commercial graphite samples	Freeze-thaw processing can be expensive and is hard to scale up	Graphene flakes enhanced thermal conductivity and heat transfer coefficients of ethylene glycol fluids by 10.5% and 30%, respectively	[183]
Manual disassembly of graphite from LIBs, washing with NMP, ethanol, and deionized water, and shear mixing for 4 h	33%–36% graphene productivity ($<$ 4 times higher than that of pristine graphite)	NMP is both expensive and toxic, and graphene productivity is still relatively low from this method	Minor graphene/solution separation observed after 48 h, specific surface area of 424.9 $m^2\,g^{-1}$	[188]
Spent graphite obtained from acid leaching of black mass, modified Hummers method, and thermal reduction at 500°C for 4 h under Ar	First work to use black mass as the feedstock, Hummers' method is found to help further purify spent graphite, and processing one tonne of spent graphite yields a profit of \$52,651.4	Reduction under Ar at an industrial scale is unrealistic	C/O ratio of 1.99, $<$ 100 ppm other impurities, I_D/I_G ratio of 1.52, and a disordered few-layer structure exhibited	[189]

The structural relationship between graphite and graphene presents unique opportunities for circular economy approaches. Specifically, spent graphite from LIBs is emerging as a more useful precursor for producing graphene compared to virgin graphite. Both materials can also be sourced from waste, further diversifying the supply of these materials and helping to balance production. The prospects for "green" graphite are promising, with several companies already scaling up production in this area. While recycled graphite is still in the early stages of commercialization, it holds significant potential for integration into current production methods for battery-grade graphite. Political factors have also played a role in driving the creation of closed-loop supply chains for critical materials, especially in Europe and the United States. Although green graphene production is a growing field, it is less critical for LIBs than graphite, due to the smaller quantities needed. Nevertheless, it continues to show promise for improving battery performance and is supported by ongoing academic research.

The transition to green graphite and graphene production faces three critical challenges that must be addressed to enable sustainable lithium-ion battery (LIB) manufacturing:

1. Process optimization –

   Both graphite spheroidization (critical for anode materials) and top-down graphene synthesis suffer from suboptimal yields. Enhancing process efficiency is essential to reduce waste and improve economic viability.

2. Eco-friendly production at scale –

   Current industrial production relies heavily on environmentally damaging methods, including high-temperature pyrolysis and hydrofluoric acid (HF) treatment. Developing scalable alternatives that maintain material quality while eliminating these hazardous processes remains a key obstacle.

3. Circular economy integration –

   While recycled graphite shows promise as a raw material for battery-grade graphite and graphene, establishing reliable, large-scale supply chains remains challenging. Wider adoption requires both technological advances and market incentives.

Addressing these challenges would significantly enhance the environmental sustainability of lithium-ion battery (LIB) manufacturing, contributing to global net-zero emissions targets. Their resolution requires a multidisciplinary approach combining: (1) Technological innovation: advancements in materials engineering and process design; (2) Economic incentives: investment mechanisms to support industrial scaling; (3) Policy frameworks: regulatory measures (e.g., recycled content mandates) to accelerate industry adoption. For instance, requiring recycled graphite in electric vehicle batteries would create immediate market demand, driving innovation in recycling technologies and making closed-loop graphite production commercially feasible. The integration of technological, economic, and policy solutions presents a robust framework for achieving environmentally responsible scale-up of these critical battery materials. These advancements will be critical for establishing truly circular and sustainable supply chains to support the growing global battery industry while meeting climate objectives.

Acknowledgments

Benjamin Robinson gratefully acknowledges the support of a PhD studentship jointly funded by Graphene Star Ltd and the University of Warwick, through the Warwick Centre for Doctoral Training in Analytical Science. This studentship supports the project "Green Graphite and Graphene for Lithium-ion Battery Innovation: Materials, Manufacturing and Characterization" (2022–2026). Activities in the manuscript have been partially supported by UK Research and Innovation (UKRI) through the Innovate UK programmes: "CONDUCTOR: Thin and Lightweight Current Collector for Lithium-ion Batteries" (Ref: 10047927) and "CONTACT: Coated Current Collector for Battery Performance Improvement" (Ref: 10041084).

Additional funding was provided by the European Union's Horizon Europe research and innovation program under the project "STREAMS: Sustainable Technologies for Reducing Europe's Battery Raw Materials" (Grant Agreement No. 101137771). All funding has supported Professor Low's leadership of the Warwick Electrochemical Engineering R&D at the University of Warwick and its R&D Programmes, contributing to the battery technology value chain. Access to facilities at the WMG Energy Innovation Centre, including the full-scale battery prototyping resources provided by the High Value Manufacturing Catapult at Warwick, is also gratefully acknowledged.

Conflicts of Interest

Sergey Alekseev is affiliated with Graphene Star Limited, United Kingdom (www.graphene-star.com), which holds two patents relevant to the subject matter of this manuscript: (a) GB2568499—A Method for Producing Graphene (published May 2019), and (b) GB2615349—Hydrodynamic Method for Processing Natural Graphite (published August 2023). The remaining authors declare no conflicts of interest.

References

1. B. Dunn, H. Kamath, and J. M. Tarascon, "Electrical Energy Storage for the Grid: A Battery of Choices," *Science* 334, no. 6058 (2011): 928–935.

2. J. Asenbauer, T. Eisenmann, M. Kuenzel, et al., "The Success Story of Graphite as a Lithium-Ion Anode Material—Fundamentals, Remaining Challenges, and Recent Developments Including Silicon (Oxide) Composites," *Sustainable Energy & Fuels* 4, no. 11 (2020): 5387–5416.

3. C. Xu, Q. Dai, L. Gaines, M. Hu, A. Tukker, and B. Steubing, "Future Material Demand for Automotive Lithium-Based Batteries," *Communications Materials* 1, no. 99 (2020): 99.

4. H. Hao, R. Tan, C. Ye, and C. T. J. Low, "Carbon-Coated Current Collectors in Lithium-Ion Batteries and Supercapacitors: Materials, Manufacture and Applications," *Carbon Energy* 6, no. 12 (2024): e604.

5. M. Zhang, R. Tan, M. Wang, Z. Zhang, C. John Low, and Y. Lai, "Hypercrosslinked Porous and Coordination Polymer Materials for Electrolyte Membranes in Lithium-Metal Batteries," *Battery Energy* 3, no. 2 (2024): 20230050.

6. J. Lai, R. Tan, H. Jiang, et al., "Development of an In Situ Polymerized Artificial Layer for Dendrite-Free and Stable Lithium Metal Batteries," *Battery Energy* 3, no. 4 (2024): 20230070.

7. S. Gifford, *The UK: A Low Carbon Location to Manufacture, Drive and Recycle Electric Vehicles* (Faraday Institution, 2021).

8. D. Yu, Z. Huang, B. Makuza, X. Guo, and Q. Tian, "Pretreatment Options for the Recycling of Spent Lithium-Ion Batteries: A Comprehensive Review," *Minerals Engineering* 173 (2021): 107218.

9. K. Persson, V. A. Sethuraman, L. J. Hardwick, et al., “Lithium Diffusion in Graphitic Carbon,” *Journal of Physical Chemistry Letters* 1, no. 8 (2010): 1176–1180.

10. L. Zhao, B. Ding, X. Y. Qin, et al., “Revisiting the Roles of Natural Graphite in Ongoing Lithium-Ion Batteries,” *Advanced Materials* 34, no. 18 (2022): 2106704.

11. A. Senyshyn, M. J. Mühlbauer, O. Dolotko, and H. Ehrenberg, “Low-Temperature Performance of Li-Ion Batteries: The Behavior of Lithiated Graphite,” *Journal of Power Sources* 282 (2015): 235–240.

12. M. Zhu, J. Yang, Z. Yu, H. Chen, and F. Pan, “Novel Hybrid Si Nanocrystals Embedded in a Conductive SiO_x@C Matrix From One Single Precursor as a High Performance Anode Material for Lithium-Ion Batteries,” *Journal of Materials Chemistry A* 5, no. 15 (2017): 7026–7034.

13. J. Wu, Y. Cao, H. Zhao, J. Mao, and Z. Guo, “The Critical Role of Carbon in Marrying Silicon and Graphite Anodes for High-Energy Lithium-Ion Batteries,” *Carbon Energy* 1, no. 1 (2019): 57–76.

14. “Critical Raw Materials Act—Consilium,” Council of the EU and the European Council, accessed April 3, 2025, https://www.consilium.europa.eu/en/infographics/critical-raw-materials/.

15. “Resilience for the Future: The UK's Critical Minerals Strategy—GOV.UK,” Department for Business, Energy & Industrial Strategy, updated March 13, 2023, accessed April 3, 2025, https://www.gov.uk/government/publications/uk-critical-mineral-strategy/resilience-for-the-future-the-uks-critical-minerals-strategy.

16. K. Tsuji, *Global Value Chains: Graphite in Lithium-Ion Batteries for Electric Vehicles* (Office of Industries, 2022).

17. A. K. Geim and K. S. Novoselov, “The Rise of Graphene,” *Nature Materials* 6, no. 3 (2007): 183–191.

18. G. Kucinskis, G. Bajars, and J. Kleperis, “Graphene in Lithium Ion Battery Cathode Materials: A Review,” *Journal of Power Sources* 240 (2013): 66–79.

19. J. Zhu, R. Duan, S. Zhang, et al., “The Application of Graphene in Lithium Ion Battery Electrode Materials,” *Journal of the Korean Physical Society* 3, no. 1 (2014): 1–8.

20. F. Y. Su, C. You, Y. B. He, et al., “Flexible and Planar Graphene Conductive Additives for Lithium-Ion Batteries,” *Journal of Materials Chemistry* 20, no. 43 (2010): 9644–9650.

21. S. A. Alekseev, A. S. Dmitriev, A. A. Dmitriev, P. G. Makarov, and I. A. Mikhailova, “Functional Energy Nanocomposites Surfaces Based on Mesoscopic Microspheres, Polymers and Graphene Flakes,” *Journal of Physics: Conference Series* 891, no. 1 (2017): 012361.

22. J. Bi, Z. Du, J. Sun, et al., “On the Road to the Frontiers of Lithium-Ion Batteries: A Review and Outlook of Graphene Anodes,” *Advanced Materials* 35, no. 16 (2023): 2210734.

23. K. Lee, H. Zhou, M. Zuba, et al., “Complex Defect Chemistry of Hydrothermally-Synthesized Nb-Substituted β'-$LiVOPO_4$,” *Journal of Materials Chemistry A* 11, no. 20 (2023): 10834–10849.

24. N. Sick, O. Krätzig, G. G. Eshetu, and E. Figgemeier, “A Review of the Publication and Patent Landscape of Anode Materials for Lithium Ion Batteries,” *Journal of Energy Storage* 43 (2021): 103231.

25. T. Schmaltz, L. Wormer, U. Schmoch, H. Döscher, et al., “Graphene Roadmap Briefs (No. 3): Meta-Market Analysis 2023,” *2D Materials* 11, no. 2 (2024): 022002.

26. M. Phadatare, R. Patil, and H. Olin, "Silicon-Nanographite Aerogel-Based Anodes for Batteries," US2022336798A1, October 20, 2022.

27. S. Laine, “Methods, Devices and Systems for Processing and Filtering Carbonaceous Compositions,” US20190060798A1, August 29, 2019.

28. W. Zhao, C. Zhao, H. Wu, L. Li, and C. Zhang, “Progress, Challenge and Perspective of Graphite-Based Anode Materials for Lithium Batteries: A Review,” *Journal of Energy Storage* 81 (2024): 110409.

29. M. Olutogun, A. Vanderbruggen, C. Frey, M. Rudolph, D. Bresser, and S. Passerini, “Recycled Graphite for More Sustainable Lithium-Ion Batteries,” *Carbon Energy* 6, no. 5 (2024): e483.

30. M. Bhar, S. Ghosh, S. Krishnamurthy, K. Yalamanchili, and S. K. Martha, “Electrochemical Compatibility of Graphite Anode From Spent Li-Ion Batteries: Recycled via a Greener and Sustainable Approach,” *ACS Sustainable Chemistry & Engineering* 10, no. 23 (2022): 7515–7525.

31. Z. J. Baum, R. E. Bird, X. Yu, and J. Ma, “Lithium-Ion Battery Recycling─Overview of Techniques and Trends,” *ACS Energy Letters* 7, no. 2 (2022): 712–719.

32. H. E. Kang, J. Ko, S. G. Song, and Y. S. Yoon, “Recent Progress in Utilizing Carbon Nanotubes and Graphene to Relieve Volume Expansion and Increase Electrical Conductivity of Si-Based Composite Anodes for Lithium-Ion Batteries,” *Carbon* 219 (2024): 118800.

33. H. L. Andersen, L. Djuandhi, U. Mittal, and N. Sharma, “Strategies for the Analysis of Graphite Electrode Function,” *Advanced Energy Materials* 11, no. 48 (2021): 2102693.

34. X. Jiang, Y. Chen, X. Meng, et al., “The Impact of Electrode With Carbon Materials on Safety Performance of Lithium-Ion Batteries: A Review,” *Carbon* 191 (2022): 448–470.

35. T. Carrère, U. Khalid, M. Baumann, M. Bouzidi, and B. Allard, “Carbon Footprint Assessment of Manufacturing of Synthetic Graphite Battery Anode Material for Electric Mobility Applications,” *Journal of Energy Storage* 94 (2024): 112356.

36. S. Niu, G. Zhu, K. Wu, and H. Zheng, “The Feasibility for Natural Graphite to Replace Artificial Graphite in Organic Electrolyte With Different Film-Forming Additives,” *Chinese Journal of Chemical Engineering* 56 (2023): 58–69.

37. Q. Q. Zhang, X. Z. Gong, and X. C. Meng, “Environment Impact Analysis of Natural Graphite Anode Material Production,” *Materials Science Forum* 913 (2018): 1011–1017.

38. S. W. Gao, X. Z. Gong, Y. Liu, and Q. Q. Zhang, “Energy Consumption and Carbon Emission Analysis of Natural Graphite Anode Material for Lithium Batteries,” *Materials Science Forum* 913 (2018): 985–990.

39. P. Engels, F. Cerdas, T. Dettmer, et al., “Life Cycle Assessment of Natural Graphite Production for Lithium-Ion Battery Anodes Based on Industrial Primary Data,” *Journal of Cleaner Production* 336 (2022): 130474.

40. C. E. L. Foss, A. M. Svensson, S. Sunde, and F. Vullum-Bruer, “Edge/Basal/Defect Ratios in Graphite and Their Influence on the Thermal Stability of Lithium Ion Batteries,” *Journal of Power Sources* 317 (2016): 177–183.

41. M. Mundszinger, S. Farsi, M. Rapp, U. Golla-Schindler, U. Kaiser, and M. Wachtler, “Morphology and Texture of Spheroidized Natural and Synthetic Graphites,” *Carbon* 111 (2017): 764–773.

42. J. Kirner, L. Zhang, Y. Qin, X. Su, Y. Li, and W. Lu, “Analysis of Graphite Materials for Fast-Charging Capabilities in Lithium-Ion Batteries,” *ECS Transactions* 85, no. 13 (2018): 33–44.

43. C. Mao, M. Wood, L. David, et al., “Selecting the Best Graphite for Long-Life, High-Energy Li-Ion Batteries,” *Journal of the Electrochemical Society* 165, no. 9 (2018): A1837–A1845.

44. K. Zaghib, X. Song, A. Guerfi, R. Rioux, and K. Kinoshita, “Purification Process of Natural Graphite as Anode for Li-Ion Batteries: Chemical Versus Thermal,” *Journal of Power Sources* 119–121 (2003): 8–15.

45. *BTR New Material Group Co, Ltd.* BTR, accessed February 20, 2025, https://www.btrchina.com/en/NegativeProducts/index.aspx?lcid=15.

46. N. Ohta, K. Nagaoka, K. Hoshi, S. Bitoh, and M. Inagaki, “Carbon-Coated Graphite for Anode of Lithium Ion Rechargeable Batteries: Graphite Substrates for Carbon Coating,” *Journal of Power Sources* 194, no. 2 (2009): 985–990.

47. S. L. Glazier, J. Li, A. J. Louli, J. P. Allen, and J. R. Dahn, “An Analysis of Artificial and Natural Graphite in Lithium Ion Pouch Cells Using Ultra-High Precision Coulometry, Isothermal Microcalorimetry, Gas Evolution, Long Term Cycling and Pressure Measurements,” *Journal of the Electrochemical Society* 164, no. 14 (2017): A3545–A3555.

48. *Graphite Mine Production Top Countries 2021*. Statista, accessed February 2, 2025, https://www.statista.com/statistics/267366/world-graphite-production/.

49. ”Towards CO_2 Neutrality Due to Carbon and Graphite,” European Carbon and Graphite Association, 2018, accessed September 14, 2024, https://ecga.net/sites/default/files/pdf/ecga_decarbonisation_brochure_i.pdf.

50. E. L. Johnson, M. Ericsson, and A. Löf, “The Mining Permitting Process in Selected Developed Economies,” *Land Use Policy* 131 (2023): 106762.

51. M. Wissler, “Graphite and Carbon Powders for Electrochemical Applications,” *Journal of Power Sources* 156, no. 2 (2006): 142–150.

52. G. R. Robinson Jr., J. M. Hammarstrom, and D. W. Olson, *Critical Mineral Resources of the United States—Economic and Environmental Geology and Prospects for Future Supply* (USGS Publications Warehouse, 2017).

53. Y. S. Wu, T. S. Yeh, Y. H. Lee, and Y. C. Lee, “Spheroidization Modification of Artificial Graphite Applied as Anode Materials for High Rate Lithium Ion Batteries,” *Advanced Materials Research* 201–203 (2011): 421–424.

54. M. Rapp, M. Mundszinger, S. Farsi, U. Golla-Schindler, U. Kaiser, and M. Wachtler, “Spheroidization of Graphite As Anode Material for Li-Ion Batteries: Spheroidization Process and Material Texture and Morphology,” *ECS Meeting Abstracts* MA2016–03, no. 2 (2016): 503.

55. F. Xi, S. Li, W. Ma, Z. Chen, K. Wei, and J. Wu, “A Review of Hydrometallurgy Techniques for the Removal of Impurities From Metallurgical-Grade Silicon,” *Hydrometallurgy* 201 (2021): 105553.

56. S. Huang, W. Wang, Q. Cui, W. Song, and S. Jiao, “Assessment of Spherical Graphite for Lithium-Ion Batteries: Techniques, China's Status, Production Market, and Recommended Policies for Sustainable Development,” *Advanced Sustainable Systems* 6, no. 11 (2022): 2200243.

57. Q. Dai, J. C. Kelly, L. Gaines, and M. Wang, “Life Cycle Analysis of Lithium-Ion Batteries for Automotive Applications,” *Batteries* 5, no. 2 (2019): 48.

58. J. B. Dunn, L. Gaines, M. Barnes, J. Sullivan, and M. Wang, *Material and Energy Flows in the Materials Production, Assembly, and End-of-Life Stages of the Automotive Lithium-Ion Battery Life Cycle* (U.S. Department of Energy, 2012).

59. D. A. Notter, M. Gauch, R. Widmer, et al., “Contribution of Li-Ion Batteries to the Environmental Impact of Electric Vehicles,” *Environmental Science & Technology* 44, no. 17 (2010): 6550–6556.

60. H. Predel, *Petroleum Coke, in Ullmann's Encyclopedia of Industrial Chemistry* (John Wiley & Sons, Ltd, 2006).

61. D. Surovtseva, E. Crossin, R. Pell, and L. Stamford, “Toward a Life Cycle Inventory for Graphite Production,” *Journal of Industrial Ecology* 26, no. 3 (2022): 964–979.

62. B. Moradi and G. G. Botte, “Recycling of Graphite Anodes for the Next Generation of Lithium Ion Batteries,” *Journal of Applied Electrochemistry* 46, no. 2 (2015): 123–148.

63. “Visualizing the Natural Graphite Supply Problem,” NGC, published November 18, 2021, accessed February 14, 2025, https://elements.visualcapitalist.com/visualizing-the-natural-graphite-supply-problem/.

64. N. B. Manjong, L. Usai, O. S. Burheim, and A. H. Strømman, “Life Cycle Modelling of Extraction and Processing of Battery Minerals—A Parametric Approach,” *Batteries* 7, no. 3 (2021): 57.

65. J. C. Abrego-Martinez, Y. Wang, V. Vanpeene, and L. Roué, “From Waste Graphite Fines to Revalorized Anode Material for Li-Ion Batteries,” *Carbon* 209 (2023): 118004.

66. A. Stewart, *Mineral Commodity Summaries—GRAPHITE (NATURAL)* (US Geological Survey, 2023).

67. *Global Li-Ion Battery Demand 2022–2030*. Statista, accessed February 16, 2025, https://www.statista.com/statistics/1419502/global-lithium-ion-battery-demand-forecast/.

68. F. Bonaccorso, A. Lombardo, T. Hasan, Z. Sun, L. Colombo, and A. C. Ferrari, “Production and Processing of Graphene and 2D Crystals,” *Materials Today* 15, no. 12 (2012): 564–589.

69. M. Coroş, F. Pogăcean, L. Măgeruşan, C. Socaci, and S. Pruneanu, “A Brief Overview on Synthesis and Applications of Graphene and Graphene-Based Nanomaterials,” *Frontiers of Materials Science* 13, no. 1 (2019): 23–32.

70. A. R. Urade, I. Lahiri, and K. S. Suresh, “Graphene Properties, Synthesis and Applications: A Review,” *JOM* 75, no. 3 (2022): 614–630.

71. N. Liu, Q. Tang, B. Huang, and Y. Wang, “Graphene Synthesis: Method, Exfoliation Mechanism and Large-Scale Production,” *Crystals* 12, no. 1 (2021): 25.

72. J. Hassoun, F. Bonaccorso, M. Agostini, et al., “An Advanced Lithium-Ion Battery Based on a Graphene Anode and a Lithium Iron Phosphate Cathode,” *Nano Letters* 14, no. 8 (2014): 4901–4906.

73. Y. Jia, J. Zhang, D. Kong, et al., “Practical Graphene Technologies for Electrochemical Energy Storage,” *Advanced Functional Materials* 32, no. 42 (2022): 2204272.

74. Y. Fu, J. Hansson, Y. Liu, et al., “Graphene Related Materials for Thermal Management,” *2D Materials* 7, no. 1 (2019): 012001.

75. R. Mo, X. Tan, F. Li, et al., “Tin-Graphene Tubes as Anodes for Lithium-Ion Batteries With High Volumetric and Gravimetric Energy Densities,” *Nature Communications* 11, no. 1 (2020): 1–11.

76. M. Wang, M. Tang, S. Chen, et al., “Graphene-Armored Aluminum Foil With Enhanced Anticorrosion Performance as Current Collectors for Lithium-Ion Battery,” *Advanced Materials* 29, no. 47 (2017): 1703882.

77. Q. Huang, M. J. Loveridge, R. Genieser, M. J. Lain, and R. Bhagat, “Electrochemical Evaluation and Phase-Related Impedance Studies on Silicon–Few Layer Graphene (FLG) Composite Electrode Systems,” *Scientific Reports* 8, no. 1 (2018): 1–9.

78. Y. Mu, M. Han, J. Li, J. Liang, and J. Yu, “Growing Vertical Graphene Sheets on Natural Graphite for Fast Charging Lithium-Ion Batteries,” *Carbon* 173 (2021): 477–484.

79. H. Liu, Y. Liu, and D. Zhu, “Chemical Doping of Graphene,” *Journal of Materials Chemistry* 21, no. 10 (2011): 3335–3345.

80. Z. S. Wu, W. Ren, L. Xu, F. Li, and H. M. Cheng, “Doped Graphene Sheets as Anode Materials With Superhigh Rate and Large Capacity for Lithium Ion Batteries,” *ACS Nano* 5, no. 7 (2011): 5463–5471.

81. D. Xiong, X. Li, Z. Bai, et al., “Superior Cathode Performance of Nitrogen-Doped Graphene Frameworks for Lithium Ion Batteries,” *ACS Applied Materials & Interfaces* 9, no. 12 (2017): 10643–10651.

82. X. Wang, X. Cao, L. Bourgeois, et al., “N-Doped Graphene-SnO_2 Sandwich Paper for High-Performance Lithium-Ion Batteries,” *Advanced Functional Materials* 22, no. 13 (2012): 2682–2690.

83. H. Lee, K. Paeng, and I. S. Kim, “A Review of Doping Modulation in Graphene,” *Synthetic Metals* 244 (2018): 36–47.

84. H. Wang, Z. L. Yi, F. Y. Su, et al., “The Effect of Removing the Native Passivation Film on the Electrochemical Performance of Lithium Metal Electrodes,” *Journal of Power Sources* 520 (2022): 230817.

85. T. Chi, X. Wang, L. Zeng, Z. Qin, X. Zhou, and Z. Liu, “Unraveling the Effect of Conductive Additives on Li-Ion Diffusion Using

Electrochemical Impedance Spectroscopy: A Case Study of Graphene vs Carbon Black," *Journal of the Electrochemical Society* 170, no. 4 (2023): 040515.

86. W. Ren and H. M. Cheng, "The Global Growth of Graphene," *Nature Nanotechnology* 9, no. 10 (2014): 726–730.

87. N. Kumar, R. Salehiyan, V. Chauke, et al., "Top-Down Synthesis of Graphene: A Comprehensive Review," *FlatChem* 27 (2021): 100224.

88. A. Gutiérrez-Cruz, A. R. Ruiz-Hernández, J. F. Vega-Clemente, D. G. Luna-Gazcón, and J. Campos-Delgado, ""A Review of Top-Down and Bottom-Up Synthesis Methods for the Production of Graphene, Graphene Oxide and Reduced Graphene Oxide," *Journal of Materials Science* 57, no. 31 (2022): 14543–14578.

89. D. A. C. Brownson and C. E. Banks, "The Electrochemistry of CVD Graphene: Progress and Prospects," *Physical Chemistry Chemical Physics* 14, no. 23 (2012): 8264–8281.

90. Y. Hernandez, V. Nicolosi, M. Lotya, et al., "High-Yield Production of Graphene by Liquid-Phase Exfoliation of Graphite," *Nature Nanotechnology* 3, no. 9 (2008): 563–568.

91. Y. Xu, H. Cao, Y. Xue, B. Li, and W. Cai, "Liquid-Phase Exfoliation of Graphene: An Overview on Exfoliation Media, Techniques, and Challenges," *Nanomaterials* 8, no. 11 (2018): 942.

92. K. R. Paton, E. Varrla, C. Backes, et al., "Scalable Production of Large Quantities of Defect-Free Few-Layer Graphene by Shear Exfoliation in Liquids," *Nature Materials* 13, no. 6 (2014): 624–630.

93. I. Imae, "Reduction of Graphene Oxide Using an Environmentally Friendly Method and Its Application to Energy-Related Materials," *Coatings* 11, no. 3 (2021): 297.

94. W. S. Hummers and R. E. Offeman, "Preparation of Graphitic Oxide," *Journal of the American Chemical Society* 80, no. 6 (1958): 1339.

95. H. L. Poh, F. Šaněk, A. Ambrosi, G. Zhao, Z. Sofer, and M. Pumera, "Graphenes Prepared by Staudenmaier, Hofmann and Hummers Methods With Consequent Thermal Exfoliation Exhibit Very Different Electrochemical Properties," *Nanoscale* 4, no. 11 (2012): 3515–3522.

96. S. Pei and H. M. Cheng, "The Reduction of Graphene Oxide," *Carbon* 50, no. 9 (2012): 3210–3228.

97. R. Arvidsson, D. Kushnir, B. A. Sandén, and S. Molander, "Prospective Life Cycle Assessment of Graphene Production by Ultrasonication and Chemical Reduction," *Environmental Science & Technology* 48, no. 8 (2014): 4529–4536.

98. S. Lin, S. F. Ng, and W. J. Ong, "Life Cycle Assessment of Environmental Impacts Associated With Oxidative Desulfurization of Diesel Fuels Catalyzed by Metal-Free Reduced Graphene Oxide," *Environmental Pollution* 288 (2021): 117677.

99. O. Winjobi, Q. Dai, and J. C. Kelly, *Update of Bill-of-Materials and Cathode Chemistry Addition for Lithium-Ion Batteries in GREET 2020* (Argonne National Laboratory, 2020).

100. N. von Drachenfels, J. Husmann, U. Khalid, F. Cerdas, and C. Herrmann, "Life Cycle Assessment of the Battery Cell Production: Using a Modular Material and Energy Flow Model to Assess Product and Process Innovations," *Energy Technology* 11, no. 5 (2023): 2200673.

101. F. Rosner, T. Bhagde, D. S. Slaughter, V. Zorba, and J. Stokes-Draut, "Techno-Economic and Carbon Dioxide Emission Assessment of Carbon Black Production," *Journal of Cleaner Production* 436 (2024): 140224.

102. S. Dong, Y. Song, K. Ye, et al., "Ultra-Fast, Low-Cost, and Green Regeneration of Graphite Anode Using Flash Joule Heating Method," *EcoMat* 4, no. 5 (2022): e12212.

103. J. Liang, P. Wang, Z. Liu, et al., "A Microcrystalline Soft Carbon Modified Hard Carbon Coating Enhances Cycling Stability and Initial Efficiency in Natural Graphite Anodes," *Sustainable Energy & Fuels* 9, no. 13 (2025): 3635–3645.

104. H. J. Kwon, S. W. Woo, Y. J. Lee, J. Y. Kim, and S. M. Lee, "Achieving High-Performance Spherical Natural Graphite Anode Through a Modified Carbon Coating for Lithium-Ion Batteries," *Energies* 14, no. 7 (2021): 1946.

105. F. I. Barre, R. G. Billy, F. A. Lopez, and D. B. Müller, "Limits to Graphite Supply in a Transition to a Post-Fossil Society," *Resources, Conservation And Recycling* 208 (2024): 107709.

106. R. Istrate, A. Mas-Fons, A. Beylot, et al., "Decarbonizing Lithium-Ion Battery Primary Raw Materials Supply Chain," *Joule* 8, no. 11 (2024): 2992–3016.

107. F. Ma, D. Tao, Y. Tao, and S. Liu, "An Innovative Flake Graphite Upgrading Process Based on HPGR, Stirred Grinding Mill, and Nanobubble Column Flotation," *International Journal of Mining Science and Technology* 31, no. 6 (2021): 1063–1074.

108. Y. Zhang, H. Zhu, J. Zhu, F. Min, J. Chen, and Q. Shi, "Effect of Inorganic Cations on Enhancing Graphite/Kerosene Adsorption and Reducing Carbon Emission in Graphite Flotation," *Fuel* 314 (2022): 122740.

109. Z. Peng, D. Li, W. Fang, et al., "Improved Flotation of Fine Flake Graphite Using a Modified Thickening Process," *Separations* 10, no. 5 (2023): 275.

110. Z. Tong, J. Lu, X. Hu, et al., "Ultrasound Pretreatment for Enhancing Fine and Ultrafine Flake Graphite Flotation Beneficiation," *ACS Omega* 9, no. 9 (2024): 10717–10726.

111. B. Biber, S. Sander, J. Martin, M. Wohlfahrt-Mehrens, and M. Mancini, "Improved Production Process With New Spheroidization Machine With High Efficiency and Low Energy Consumption for Rounding Natural Graphite for Li-Ion Battery Applications," *Carbon* 201 (2023): 847–855.

112. S. M. Lee, D. S. Kang, and J. S. Roh, "Bulk Graphite: Materials and Manufacturing Process," *Carbon Letters* 16, no. 3 (2015): 135–146.

113. B. P. Thapaliya, H. Luo, P. Halstenberg, H. M. Meyer, J. R. Dunlap, and S. Dai, "Low-Cost Transformation of Biomass-Derived Carbon to High-Performing Nano-Graphite via Low-Temperature Electrochemical Graphitization," *ACS Applied Materials & Interfaces* 13, no. 3 (2021): 4393–4401.

114. A. S. Kamal, R. Othman, and N. H. Jabarullah, ""Preparation and Synthesis of Synthetic Graphite From Biomass Waste: A Review," *Systematic Reviews in Pharmacy* 11, no. 2 (2020): 881–894.

115. S. Ko, Y. J. Kwon, J. U. Lee, and Y. P. Jeon, "Preparation of Synthetic Graphite From Waste PET Plastic," *Journal of Industrial and Engineering Chemistry* 83 (2020): 449–458.

116. W. E. Tenhaeff, O. Rios, K. More, and M. A. McGuire, "Highly Robust Lithium Ion Battery Anodes From Lignin: An Abundant, Renewable, and Low-Cost Material," *Advanced Functional Materials* 24, no. 1 (2014): 86–94.

117. C. Liang, Y. Chen, M. Wu, et al., "Green Synthesis of Graphite From CO_2 Without Graphitization Process of Amorphous Carbon," *Nature Communications* 12, no. 1 (2021): 1–9.

118. Z. Zhu, H. Zuo, S. Li, et al., "A Green Electrochemical Transformation of Inferior Coals to Crystalline Graphite for Stable Li-Ion Storage," *Journal of Materials Chemistry A* 7, no. 13 (2019): 7533–7540.

119. X. Zhou, F. Chen, T. Bai, et al., "Interconnected Highly Graphitic Carbon Nanosheets Derived From Wheat Stalk as High Performance Anode Materials for Lithium Ion Batteries," *Green Chemistry* 18, no. 7 (2016): 2078–2088.

120. K. Yang, Z. Zhao, X. Xin, Z. Tian, K. Peng, and Y. Lai, "Graphitic Carbon Materials Extracted From Spent Carbon Cathode of Aluminium Reduction Cell as Anodes for Lithium Ion Batteries: Converting the Hazardous Wastes into Value-Added Materials," *Journal of the Taiwan Institute of Chemical Engineers* 104 (2019): 201–209.

121. S. W. Han, D. W. Jung, J. H. Jeong, and E. S. Oh, "Effect of Pyrolysis Temperature on Carbon Obtained From Green Tea Biomass for Superior Lithium Ion Battery Anodes," *Chemical Engineering Journal* 254 (2014): 597–604.

122. N. A. Banek, K. R. McKenzie, D. T. Abele, and M. J. Wagner, "Sustainable Conversion of Biomass to Rationally Designed Lithium-Ion Battery Graphite," *Scientific Reports* 12, no. 1 (2022): 8080.

123. W. Liu, J. Liu, M. Zhu, et al., "Recycling of Lignin and Si Waste for Advanced Si/C Battery Anodes," *ACS Applied Materials & Interfaces* 12, no. 51 (2020): 57055–57063.

124. L. Xie, C. Tang, Z. Bi, et al., "Hard Carbon Anodes for Next-Generation Li-Ion Batteries: Review and Perspective," *Advanced Energy Materials* 11, no. 38 (2021): 2101650.

125. S. Wu, N. Kaden, and K. Dröder, "A Systematic Review on Lithium-Ion Battery Disassembly Processes for Efficient Recycling," *Batteries* 9, no. 6 (2023): 297.

126. N. Lin, Z. Jia, Z. Wang, et al., "Understanding the Crack Formation of Graphite Particles in Cycled Commercial Lithium-Ion Batteries by Focused Ion Beam—Scanning Electron Microscopy," *Journal of Power Sources* 365 (2017): 235–239.

127. C. P. Makwarimba, M. Tang, Y. Peng, et al., "Assessment of Recycling Methods and Processes for Lithium-Ion Batteries," *iScience* 25, no. 5 (2022): 104321.

128. Y. Yao, M. Zhu, Z. Zhao, B. Tong, Y. Fan, and Z. Hua, ""Hydrometallurgical Processes for Recycling Spent Lithium-Ion Batteries: A Critical Review," *ACS Sustainable Chemistry & Engineering* 6, no. 11 (2018): 13611–13627.

129. B. Makuza, Q. Tian, X. Guo, K. Chattopadhyay, and D. Yu, "Pyrometallurgical Options for Recycling Spent Lithium-Ion Batteries: A Comprehensive Review," *Journal of Power Sources* 491 (2021): 229622.

130. I. Rey, C. Vallejo, G. Santiago, M. Iturrondobeitia, and E. Lizundia, "Environmental Impacts of Graphite Recycling From Spent Lithium-Ion Batteries Based on Life Cycle Assessment," *ACS Sustainable Chemistry & Engineering* 9, no. 43 (2021): 14488–14501.

131. A. Sarkar, P. Shrotriya, and I. C. Nlebedim, "Electrochemical-Driven Green Recovery of Lithium, Graphite and Cathode From Lithium-Ion Batteries Using Water," *Waste Management* 150 (2022): 320–327.

132. Y. H. Xiao, J. Li, W. G. Huang, et al., "Green & Efficient Regeneration of Graphite Anode From Spent Lithium Ion Batteries Enabled by Asphalt Coating," *Journal of Materials Science: Materials in Electronics* 33, no. 21 (2022): 16740–16752.

133. H. Wang, Y. Huang, C. Huang, et al., "Reclaiming Graphite From Spent Lithium Ion Batteries Ecologically and Economically," *Electrochimica Acta* 313 (2019): 423–431.

134. C. Yuwen, B. Liu, H. Zhang, et al., "Efficient Recovery and Regeneration of Waste Graphite Through Microwave Stripping From Spent Batteries Anode for High-Performance Lithium-Ion Batteries," *Journal of Cleaner Production* 333 (2022): 130197.

135. C. Yi, Y. Yang, T. Zhang, X. Wu, W. Sun, and L. Yi, "A Green and Facile Approach for Regeneration of Graphite From Spent Lithium Ion Battery," *Journal of Cleaner Production* 277 (2020): 123585.

136. Z. Ma, Y. Zhuang, Y. Deng, et al., "From Spent Graphite to Amorphous sp^2+sp^3 Carbon-Coated sp^2 Graphite for High-Performance Lithium Ion Batteries," *Journal of Power Sources* 376 (2018): 91–99.

137. X. Ma, M. Chen, B. Chen, Z. Meng, and Y. Wang, "High-Performance Graphite Recovered From Spent Lithium-Ion Batteries," *ACS Sustainable Chemistry & Engineering* 7, no. 24 (2019): 19732–19738.

138. Y. Gao, C. Wang, J. Zhang, et al., "Graphite Recycling From the Spent Lithium-Ion Batteries by Sulfuric Acid Curing-Leaching Combined With High-Temperature Calcination," *ACS Sustainable Chemistry & Engineering* 8, no. 25 (2020): 9447–9455.

139. J. Zhang, X. Li, D. Song, Y. Miao, J. Song, and L. Zhang, "Effective Regeneration of Anode Material Recycled From Scrapped Li-Ion Batteries," *Journal of Power Sources* 390 (2018): 38–44.

140. H. Xiao, G. Ji, L. Ye, et al., "Efficient Regeneration and Reutilization of Degraded Graphite as Advanced Anode for Lithium-Ion Batteries," *Journal of Alloys and Compounds* 888 (2021): 161593.

141. Y. Lai, X. Zhu, J. Li, et al., ""Recovery and Regeneration of Anode Graphite From Spent Lithium-Ion Batteries Through Deep Eutectic Solvent Treatment: Structural Characteristics, Electrochemical Performance and Regeneration Mechanism," *Chemical Engineering Journal* 457 (2023): 141196.

142. C. Xu, G. Ma, W. Yang, et al., "One-Step Reconstruction of Acid Treated Spent Graphite for High Capacity and Fast Charging Lithium-Ion Batteries," *Electrochimica Acta* 415 (2022): 140198.

143. D. Ruan, F. Wang, L. Wu, et al., "A High-Performance Regenerated Graphite Extracted From Discarded Lithium-Ion Batteries," *New Journal of Chemistry* 45, no. 3 (2021): 1535–1540.

144. Y. He, T. Zhang, F. Wang, G. Zhang, W. Zhang, and J. Wang, "Recovery of $LiCoO_2$ and Graphite From Spent Lithium-Ion Batteries by Fenton Reagent-Assisted Flotation," *Journal of Cleaner Production* 143 (2017): 319–325.

145. Y. Yang, S. Song, S. Lei, et al., "A Process for Combination of Recycling Lithium and Regenerating Graphite From Spent Lithium-Ion Battery," *Waste Management* 85 (2019): 529–537.

146. X. Zhu, J. Xiao, Q. Mao, et al., "A Promising Regeneration of Waste Carbon Residue From Spent Lithium-Ion Batteries via Low-Temperature Fluorination Roasting and Water Leaching," *Chemical Engineering Journal* 430 (2022): 132703.

147. Y. Gao, J. Zhang, H. Jin, et al., "Regenerating Spent Graphite From Scrapped Lithium-Ion Battery by High-Temperature Treatment," *Carbon* 189 (2022): 493–502.

148. Q. Chen, L. Huang, J. Liu, Y. Luo, and Y. Chen, "A New Approach to Regenerate High-Performance Graphite From Spent Lithium-Ion Batteries," *Carbon* 189 (2022): 293–304.

149. D. Hou, Z. Guo, Y. Wang, et al., "Microwave-Assisted Reconstruction of Spent Graphite and Its Enhanced Energy-Storage Performance as LIB Anodes," *Surfaces and Interfaces* 24 (2021): 101098.

150. W. Fan, J. Zhang, R. Ma, Y. Chen, and C. Wang, "Regeneration of Graphite Anode From Spent Lithium-Ion Batteries via Microwave Calcination," *Journal of Electroanalytical Chemistry* 908 (2022): 116087.

151. E. V. Beletskii, V. V. Pakalnis, D. A. Lukyanov, D. V. Anishchenko, A. I. Volkov, and O. V. Levin, "Recycling Spent Graphite Anodes into a Graphite/Graphene Oxide Composite via Plasma Solution Treatment for Reuse in Lithium-Ion Batteries," *Journal of Environmental Chemical Engineering* 11, no. 1 (2023): 109234.

152. K. Dai, Z. Wang, G. Ai, et al., "The Transformation of Graphite Electrode Materials in Lithium-Ion Batteries After Cycling," *Journal of Power Sources* 298 (2015): 349–354.

153. K. Chen, Y. Ding, L. Yang, et al., "Recycling of Spent Lithium–Ion Battery Graphite Anodes via a Targeted Repair Scheme," *Resources, Conservation And Recycling* 201 (2024): 107326.

154. C. Yi, P. Ge, X. Wu, W. Sun, and Y. Yang, "Tailoring Carbon Chains for Repairing Graphite From Spent Lithium-Ion Battery Toward Closed-Circuit Recycling," *Journal of Energy Chemistry* 72 (2022): 97–107.

155. S. S. Zhang, "Is Li/Graphite Half-Cell Suitable for Evaluating Lithiation Rate Capability of Graphite Electrode?," *Journal of the Electrochemical Society* 167, no. 10 (2020): 100510.

156. T. Hou, G. Yang, N. N. Rajput, et al., "The Influence of FEC on the Solvation Structure and Reduction Reaction of $LiPF_6$/EC Electrolytes and Its Implication for Solid Electrolyte Interphase Formation," *Nano Energy* 64 (2019): 103881.

157. J. Tan, J. Matz, P. Dong, J. Shen, and M. Ye, “A Growing Appreciation for the Role of LiF in the Solid Electrolyte Interphase,” *Advanced Energy Materials* 11, no. 16 (2021): 2100046.

158. J. Munuera, L. Britnell, C. Santoro, R. Cuéllar-Franca, and C. Casiraghi, “A Review on Sustainable Production of Graphene and Related Life Cycle Assessment,” *2D Materials* 9, no. 1 (2021): 012002.

159. S. Zhao, S. Xie, Z. Zhao, J. Zhang, L. Li, and Z. Xin, “Green and High-Efficiency Production of Graphene by Tannic Acid-Assisted Exfoliation of Graphite in Water,” *ACS Sustainable Chemistry & Engineering* 6, no. 6 (2018): 7652–7661.

160. B. Gürünlü, Ç. Taşdelen Yücedağ, and M. R. Bayramoğlu, “Green Synthesis of Graphene From Graphite in Molten Salt Medium,” *Journal of Nanomaterials* 2020 (2020): 7029601.

161. X. Chen, Y. Zhu, W. Peng, et al., “Direct Exfoliation of the Anode Graphite of Used Li-Ion Batteries into Few-Layer Graphene Sheets: A Green and High Yield Route to High-Quality Graphene Preparation,” *Journal of Materials Chemistry A* 5, no. 12 (2017): 5880–5885.

162. P.-C. Lin, J.-Y. Wu, and W.-R. Liu, “Green and Facile Synthesis of Few-Layer Graphene via Liquid Exfoliation Process for Lithium-Ion Batteries,” *Scientific Reports* 8, no. 9766 (2018.

163. A. R. Kamali, “Eco-Friendly Production of High Quality Low Cost Graphene and Its Application in Lithium Ion Batteries,” *Green Chemistry* 18, no. 7 (2016): 1952–1964.

164. L. Yao, W. Cao, J. Zhao, et al., “Regulating Bifunctional Flower-Like $NiFe_2O_4$/Graphene for Green EMI Shielding and Lithium Ion Storage,” *Journal of Materials Science & Technology* 127 (2022): 48–60.

165. H. Xu, Y. Wang, R. Chen, et al., “A Green-Synthetic Spiderweb-Like Si@Graphene-Oxide Anode Material With Multifunctional Citric Acid Binder for High Energy-Density Li-Ion Batteries,” *Carbon* 157 (2020): 330–339.

166. R. Zhang, D. Wang, L. C. Qin, et al., “$MnCO_3$/Mn_3O_4/Reduced Graphene Oxide Ternary Anode Materials for Lithium-Ion Batteries: Facile Green Synthesis and Enhanced Electrochemical Performance,” *Journal of Materials Chemistry A* 5, no. 32 (2017): 17001–17011.

167. L. Y. Beaulieu, K. W. Eberman, R. L. Turner, L. J. Krause, and J. R. Dahn, “Colossal Reversible Volume Changes in Lithium Alloys,” *Electrochemical and Solid-State Letters* 4, no. 9 (2001): A137–A140.

168. P. Barai, A. Mistry, and P. P. Mukherjee, “Poromechanical Effect in the Lithium–Sulfur Battery Cathode,” *Extreme Mechanics Letters* 9 (2016): 359–370.

169. S. Chen, P. Bao, X. Huang, B. Sun, and G. Wang, “Hierarchical 3D Mesoporous Silicon@Graphene Nanoarchitectures for Lithium Ion Batteries With Superior Performance,” *Nano Research* 7, no. 1 (2014): 85–94.

170. B. Papandrea, X. Xu, Y. Xu, et al., “Three-Dimensional Graphene Framework With Ultra-High Sulfur Content for a Robust Lithium–Sulfur Battery,” *Nano Research* 9, no. 1 (2016): 240–248.

171. I. H. Son, J. H. Park, S. Park, et al., “Graphene Balls for Lithium Rechargeable Batteries With Fast Charging and High Volumetric Energy Densities,” *Nature Communications* 8, no. 1 (2017): 1561.

172. Z. Wang, S. Xia, and P. Wang, “Conductive Adhesive for Lithium-Ion Battery and Preparation Method Therefor, Lithium-Ion Battery Electrode Plate and Preparation Method Therefor, and Lithium-Ion Battery,” EP3444881B1, December 7, 2016.

173. H. P. Jampani, M. W. Stowell, A. Kumar, et al., “Advanced Lithium (Li) Ion and Lithium Sulfur (LiS) Batteries,” US20210126244A1, September 27, 2021.

174. P. J. Risby and D. Pennington, “Method for Processing a Gas and a Device for Performing the Method,” CA2833965C, July 1, 2019.

175. J. P. Romero, G. S. Abellán, E. M. Coronado, et al., “Device for Storing and/or Converting Energy and Method for Manufacturing Such a Device,” WO2024121442A1, June 13, 2024.

176. J. Holman, *FEATURE: Graphite Supply a Concern in Meeting Growing Battery Demand*. S&P Global Commodity Insights, accessed August 1, 2024, https://www.spglobal.com/commodityinsights/en/market-insights/latest-news/energy-transition/021622-feature-graphite-supply-a-concern-in-meeting-growing-battery-demand.

177. *Electricity Price by Country 2023*. Statista, accessed August 1, 2024, https://www.statista.com/statistics/263492/electricity-prices-in-selected-countries/.

178. A. Vanderbruggen, N. Hayagan, K. Bachmann, et al., “Lithium-Ion Battery Recycling─Influence of Recycling Processes on Component Liberation and Flotation Separation Efficiency,” *ACS ES&T Engineering* 2, no. 11 (2022): 2130–2141.

179. A. M. Salces, I. Bremerstein, M. Rudolph, and A. Vanderbruggen, “Joint Recovery of Graphite and Lithium Metal Oxides From Spent Lithium-Ion Batteries Using Froth Flotation and Investigation on Process Water Re-Use,” *Minerals Engineering* 184 (2022): 107670.

180. A. Vanderbruggen, A. Salces, A. Ferreira, M. Rudolph, and R. Serna-Guerrero, “Improving Separation Efficiency in End-of-Life Lithium-Ion Batteries Flotation Using Attrition Pre-Treatment,” *Minerals* 12, no. 1 (2022): 72.

181. A. Vanderbruggen, J. Sygusch, M. Rudolph, and R. Serna-Guerrero, “A Contribution to Understanding the Flotation Behavior of Lithium Metal Oxides and Spheroidized Graphite for Lithium-Ion Battery Recycling,” *Colloids and Surfaces, A: Physicochemical and Engineering Aspects* 626 (2021): 127111.

182. W. Mrozik, M. A. Rajaeifar, O. Heidrich, and P. Christensen, “Environmental Impacts, Pollution Sources and Pathways of Spent Lithium-Ion Batteries,” *Energy & Environmental Science* 14, no. 14 (2021): 6099–6121.

183. Y. Qiao, W. Sheng, C. He, et al., “A Facile Freeze–Thaw Ultrasonic Assisted Circulation Method of Graphite Flakes Prepared by Anode Graphite From Spent Lithium-Ion Batteries for Application in Nanofluids,” *Sustainable Energy & Fuels* 5, no. 19 (2021): 4882–4894.

184. K. He, Z. Y. Zhang, and F. S. Zhang, “Synthesis of Graphene and Recovery of Lithium From Lithiated Graphite of Spent Li-Ion Battery,” *Waste Management* 124 (2021): 283–292.

185. J. Yu, M. Lin, Q. Tan, and J. Li, “High-Value Utilization of Graphite Electrodes in Spent Lithium-Ion Batteries: From 3D Waste Graphite to 2D Graphene Oxide,” *Journal of Hazardous Materials* 401 (2021): 123715.

186. W. Zhang, Z. Liu, J. Xia, et al., “Preparing Graphene From Anode Graphite of Spent Lithium-Ion Batteries,” *Frontiers of Environmental Science and Engineering* 11, no. 5 (2017): 1–8.

187. L. Zhao, X. Liu, C. Wan, X. Ye, and F. Wu, “Soluble Graphene Nanosheets From Recycled Graphite of Spent Lithium Ion Batteries,” *Journal of Materials Engineering and Performance* 27, no. 2 (2018): 875–880.

188. Y. Zhang, N. Song, J. He, R. Chen, and X. Li, “Lithiation-Aided Conversion of End-of-Life Lithium-Ion Battery Anodes to High-Quality Graphene and Graphene Oxide,” *Nano Letters* 19, no. 1 (2019): 512–519.

189. X. Xie, J. Zhang, Y. Chen, and C. Wang, “A Method for the Preparation of Graphene From Spent Graphite of Retired Lithium-Ion Batteries,” *Journal of Power Sources* 594 (2024): 234023.

190. J. Zhu, D. Yang, Z. Yin, Q. Yan, and H. Zhang, “Graphene and Graphene-Based Materials for Energy Storage Applications,” *Small* 10, no. 17 (2014): 3480–3498.

191. A. Sobianowska-Turek, W. Urbańska, A. Janicka, M. Zawiślak, and J. Matla, "The Necessity of Recycling of Waste Li-Ion Batteries Used in Electric Vehicles as Objects Posing a Threat to Human Health and the Environment," *Recycling* 6, no. 2 (2021): 35.

192. Z. Li, J. Li, and L. Wang, "Renewed Graphite for High-Performance Lithium-Ion Batteries: Catalytic Graphitization Approach," *Journal of Materials Science: Materials in Electronics* 35, no. 8 (2024): 1–16.

193. J. Liu, H. Shi, X. Hu, et al., "Critical Strategies for Recycling Process of Graphite From Spent Lithium-Ion Batteries: A Review," *Science of the Total Environment* 816 (2022): 151621.

194. J. Paquet, *Life Cycle Assessment of NMG's Graphite Advanced Materials Confirm Minimal Environmental Footprint* (Nouveau Monde Graphite, 2022).

Biographies

Benjamin Robinson is a PhD researcher in the Analytical Sciences Centre for Doctoral Training at the University of Warwick. His research focuses on developing and optimising low-energy processing routes to produce battery-grade graphite/graphene with improved electrochemical performance. He also investigates graphite–silicon composite anodes to enhance energy density in next-generation lithium-ion batteries (LIBs). Beyond fundamental research, he has contributed to multiple battery scale-up projects on the Warwick Manufacturing Group's pilot line and is actively exploring sustainable recycling approaches for end-of-life LIBs. He earned his BSc (Hons) in Chemistry from the University of Warwick in 2021 before commencing his PhD within the Warwick Electrochemical Engineering Group.

Dr. Jie Yang is an assistant professor in Warwick Manufacturing Group at the University of Warwick. He received his PhD degree in Chemistry at the University of Manchester. He previously worked as a research fellow at the National University of Singapore. His research interests mainly focus on developing advanced energy storage devices (i.e., batteries and supercapacitors) based on new energy materials and advanced manufacturing technology, including the synthesis of novel materials (electrode materials, solid electrolytes, additives), electrode preparation techniques (wet process, dry process, electrochemical methods), exploration of energy storage mechanisms, commercialization feasibility assessment of new materials and recycling of critical materials.

Dr. Chee Tong John Low is an associate professor at the University of Warwick, who leads the Warwick Electrochemical Engineering R&D team, focusing on advancing industrial priorities in developing, manufacturing, and recycling electrochemical power devices such as automotive batteries, supercapacitors, flow batteries, and fuel cells. With expertise in chemical engineering and electrochemistry, he drives programmes that de-risk manufacturing innovation and accelerate technology readiness. He has delivered major national (EPSRC, Innovate UK), international funding (Horizon Europe), and industrial contracts that advance commercialization and patent development. He integrates research and education that address global energy storage and metal circularity challenges, through WMG Skills Centre and WMG Energy Innovation Centre.