

Environmental Impacts of Graphite Recycling from Spent Lithium-Ion Batteries Based on Life Cycle Assessment

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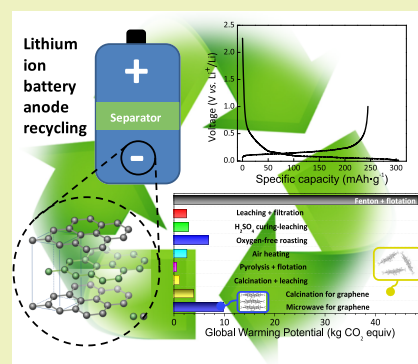
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Supporting Information

ABSTRACT: With the emergence of portable electronics and electric vehicle adoption, the last decade has witnessed an increasing fabrication of lithium-ion batteries (LIBs). The future development of LIBs is threatened by the limited reserves of virgin materials, while the inadequate management of spent batteries endangers environmental and human health. According to the Circular Economy principles aiming at reintroducing end-of-life materials back into the economic cycle, further attention should be directed to the development and implementation of battery recycling processes. To enable sustainable paths for graphite recovery, the environmental footprint of state-of-the-art graphite recycling through life cycle assessment is analyzed quantifying the contribution of nine recycling methods combining pyrometallurgical and hydrometallurgical approaches to indicators such as global warming, ozone layer depletion potential, ecotoxicity, eutrophication, or acidification. Laboratory-scale recycling is scaled up into pilot-scale processes able to treat 100 kg of spent graphite. With values ranging from 0.53 to 9.76 kg-CO₂ equiv. per 1 kg of graphite, energy consumption and waste acid generation are the main environmental drivers. A sensitivity analysis demonstrates a 20–73% impact reduction by limiting to one-fourth the amount of H₂SO₄. Combined processes involving hydrometallurgy and pyrometallurgy give environmentally preferable results. The electrochemical performance of regenerated graphite is also compared with virgin battery-grade graphite. This work provides cues boosting the environmentally sustainable recycling of spent graphite from lithium-ion batteries, strengthening the implementation of circular approaches in the battery industry.

KEYWORDS: lithium-ion battery, recycling, anode, graphite, life cycle assessment, environmental impact, ecodesign, circular economy



INTRODUCTION

Since their commercialization in the early 90s, the demand for lithium-ion batteries (LIBs) has increased exponentially.¹ Although they were initially applied into portable electronics, the last decade has witnessed an increasing use of LIBs into electric vehicles provided their high specific capacity, adequate energy density, no memory effect, low self-discharge, and acceptable cycling performance over conventional secondary batteries.² LIBs are composed by two electrode materials, where Li⁺ ions are intercalated back and forth in a reversible way, delivering an electrical power to the external circuit.³ During the charge, the oxidation of the positive electrode material (cathode) and the lithiation of the negative electrode material (anode) occur, while upon discharge, Li⁺ is extracted from the anode to be inserted into the cathode.⁴ Among the different materials applied as cathodes, LiCoO₂, LiMn₂O₄, LiFePO₄, or the more recent LiNi_xMn_yCo_zO₂ and LiNi_xCo_yAl_zO₂ are the most relevant ones.⁵ From the other side, carbonaceous materials have been predominantly used as the anode material because they are electrochemically stable and relatively cheap/widely available.^{6–8} When Li⁺ is intercalated into the graphite layers, Li_xC₆ is formed, yielding a maximum theoretical capacity of 372 mA h g⁻¹ for $x = 1$. Additionally, a

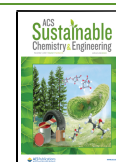
porous separator soaked into a liquid electrolyte (or a gel-like or solid electrolyte) ensures an electrical insulation between the anode and the cathode while promoting efficient ion transport between them.⁹ Typically, LIBs use a lithium salt such as LiPF₆ dissolved into a mixture of organic solvents as the electrolyte, while a polymeric membrane acts as a separator.

Unfortunately, the development of LIBs is threatened by the increasing cost and limited reserves of virgin materials used in the battery industry.¹⁰ The global demand of LIBs has escalated even more due to the accelerated electric vehicle adoption during the COVID-19 pandemic.¹¹ This vast demand encompasses great challenges related to the exhaustion of nonrenewable resources and the management of spent batteries. The recovery/recycling of LIBs has been mainly concentrated on the most economically valuable materials, that

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is, cobalt, lithium, nickel, manganese, copper, and aluminum.¹² However, the commercial breakthrough of LIBs resulted in the inclusion of materials with lower added value, such as natural graphite, in the 2020 list of the European Union (EU) of critical raw materials (CRMs), leaving no doubt that the supply chain of natural graphite is also at risk.^{7,13} Indeed, the mass of graphite takes around 15–20% by weight of batteries powering electric vehicles, accounting for nearly ~10% of the economic value of the batteries.¹⁴ As a result, as for 2020, nearly 456,000 tons of graphite were used for LIBs, with China producing 69% of the global supply of natural graphite.¹⁵ This considerable amount of hazardous solid waste containing heavy metals, toxic and flammable electrolytes, and diverse organic matters will require extensive landfill resources and could threaten human health and the ecological environment if not handled properly. Indeed, spent batteries are the fastest growing type of waste electrical and electronic equipment (WEEE).¹⁶

In line with the Circular Economy principles aiming at reintroducing end-of-life materials back into the economic cycle,¹⁷ the recycling and reuse of spent graphite from batteries are an urgent task. This priority is reflected by the increasing regulations mandating that LIBs should enter the Circular Economy instead of being discarded (“Sustainable Batteries Regulation” from the European Commission or “Used Lithium-Ion Batteries” from the United States Environmental Protection Agency). Although closing the loops through reuse, repair, refurbish, or remanufacture approaches is generally preferred,¹⁸ in the near term, a graphite recycling route is to be favored over second-life applications.¹⁹ *A priori*, recycling offers clear environmental benefits, such as improved resource efficiency, lower carbon emissions, and reductions in the amount of waste. Taking into account that by 2030, nearly 300 million of electric vehicles are expected to be used globally,¹⁹ providing robust and environmentally friendly anode recycling approaches is an urgent task.

Spent graphite typically presents undesired metal impurities (Li, Al, Co, Cu, Ni, Fe, and Mn), organic electrolytes, and polymeric binders as a result of battery recycling processes involving mechanical crushing and sorting processes to extract the cathode materials.^{12,20} Moreover, the structure of graphite is usually damaged during battery operation due to charging/discharging cycles (Li^+ insertion/extraction).²¹ Currently, spent graphite is abandoned or incinerated, encompassing severe environmental risks including particulate contamination and the emission of greenhouse gases (GHGs).²² For effective recycling, spent graphite needs to be also regenerated. Graphite recycling/regeneration has been accomplished through diverse approaches, including hydrometallurgical methods based on acid–base leaching processes (for example by using acids HCl or H_2SO_4)^{21,23} or a pyrometallurgical process where graphite is treated under temperatures above 1000 °C so that the residual metals, metal oxides, and binders are gasified and the graphite structure is repaired.²⁴ In the former, the use of inorganic acids encompasses serious environmental human and environmental safety issues,²⁵ while large amounts of energy are required to power smelting furnaces in the latter. Some companies such as Umicore are applying the pyrometallurgical process to recover high-value metals from batteries.²⁶

Most of the examples of industrial recycling of LIBs combine pyrometallurgical and hydrometallurgical unit operations. Spent batteries can be coprocessed in existing primary or

secondary Co, Cu, and Ni smelters or into specifically dedicated plants. Nickelhütte Aue GmbH (Germany) and Glencore Nikkelverk AS (Norway) are relevant examples in the first group, while Umicore SA (Belgium) is an illustrative example of a recycling plant specifically designed for batteries (nickel metal hydride batteries and LIBs).^{27,28} With an installed capacity of 7000 metric tons-year⁻¹, Umicore SA proceeds with a first pyrometallurgical phase (1450 °C) to convert batteries into a Co/Ni/Cu alloy, a Li-bearing slag, and a fly ash phase. In the subsequent hydrometallurgical process, the alloy is refined to convert the metals into active cathode materials for new batteries. Although *a priori*, the hydrometallurgical treatment can enable the recovery of other materials such as Mn or graphite from the so-called black mass, little attention has been paid to recover graphite *via* industrial processes, and the recovery is focused on transition metals (mostly due to economic reasons).²⁹ In this sense, a recent patent has demonstrated the potential recovery of graphite from spent LIBs (16 wt % of the battery).³⁰ However and despite the urgency for developing sustainable recycling routes, little efforts have been devoted to the assessment and public disclosure of the environmental impacts of these industrial processes.

However, it remains unclear whether hydrometallurgical or pyrometallurgical methods are environmentally preferred. In this context, life cycle assessment (LCA) offers the means to quantify the environmental burdens of recycling processes.³¹ LCA can be applied to batteries to determine the full environmental sustainability by analyzing the contribution of recycling processes to indicators such as global warming, ozone layer depletion potential, ecotoxicity, eutrophication, or acidification that can be computed.³² Having accurate information on the potential environmental impact regarding batteries is considered of prime interest to set the scene for future optimization under ecodesign guidelines. As summarized in Scheme 1, this work sheds light on the environmental

Scheme 1. Schematic Representation Showing the Life Cycle of Lithium-Ion Batteries with a Special Emphasis on the Recycling Phase



impacts associated with nine state-of-the-art graphite recycling methods to enable future design of battery recycling with reduced environmental impacts, a priority for initiatives such as Batteries Europe.³³ The pilot-scale environmental impacts of LIB anode (graphite) recycling and regeneration approaches are quantified, and a sensitivity analysis is performed to explore environmentally friendlier scenarios. To deepen the understanding on the recycling and regeneration processes, the electrochemical performance of resulting graphite is discussed. This work aims to serve as a road map to boost environ-

mentally sustainable recycling and reuse of graphite from lithium-ion batteries to strengthen the implementation of Circular Economy approaches in the battery industry.

EXPERIMENTAL SECTION

Goal, Scope, Battery Selection, and Inventory Analysis. The environmental impacts of nine graphite recycling processes were quantified and compared using a life cycle assessment methodology. These recycling processes were selected as representative state-of-the-art approaches from recently published investigations and involved a combination of hydrometallurgical and pyrometallurgical approaches. In comparison with traditional cathode recycling approaches focused to reclaim individual materials from the discarded compound (Li, Co, Ni, or Mn), most of the LIB anode recycling approaches were aimed at the reuse of spent graphite in new batteries. Accordingly, seven of the works focused on the regeneration of spent graphite so that it can be applied again in a secondary battery, while two works used graphite as a platform material to synthesize value-added materials such as graphene oxide.

To provide a global scope, the obtained environmental impacts were grouped into 18 midpoint impact categories according to the ReCiPe 2016 Midpoint method. This method assumed that the impacts from future extractions were included in the analysis.³⁴ The global warming potential (GWP) (kg-CO₂ equiv.) was used to compare the environmental impacts among different processes and stages.

Generally speaking, spent LIBs were first discharged to avoid safety hazards during crushing stages in recycling processes and during storage and transportation. The electrochemical discharge is typically carried out by immersion of spent batteries in solutions such as 5 wt % NaCl aqueous solution.³⁵ Second, a pretreatment involving mechanical separation processes was applied to spent batteries to treat the external casing and dismantle metallic and plastic parts. Batteries were then cut, shredded, and sieved to obtain a powder including cathode materials, anode materials, and other impurities. Our analysis started here (if not otherwise indicated), where graphite was already separated from the other battery components. This allowed leaving aside of previous steps of battery handling to focus solely on the environmental impacts of the recycling/regeneration process, so the obtained results were meaningful to carefully analyze the impacts of the recycling/regeneration itself. We estimated that this approach allowed obtaining greater accuracy when comparing the results. Accordingly, the scope of the study is schematically shown in Figure 1.

Details on the inventory modeling showing specific flowcharts of the recycling process (Figures S1–S9), process calculation, and extended inventory data (Tables S1–S9) are provided in the Supporting Information. The summarized material and energy input inventory of the anode recycling/regeneration processes is shown in Table 1 (Table S10 shows the full output list corresponding to the pyrolysis + flotation process).

As laboratory-scale LCA has been demonstrated to be an effective advisory tool for emerging technologies,³⁶ we extracted the inventory modeling from laboratory-scale studies to demonstrate the feasibility of different recycling processes and guide their future industrial-scale implementation. This assessment relied on secondary data (obtained from published manuscripts) that estimates the inputs and outputs of different graphite recycling methods. When required, the original authors of the specific studies were contacted to get further experimental details. The constructed recycling model was based on stoichiometric calculations from laboratory-scale to pilot-scale recycling processes. Accordingly, the amount of the reagents used was proportional to that reported for laboratory-scale processes (typically in the range of tens to hundreds of grams) but increased to reach an input of 100 kg of graphite. To scale up the energy demand, we used the energy requirements of pilot-scale equipment, including stirrers, centrifuges, vacuum filtration instruments, furnaces, and so on. Reaction volumes with a security factor of 2 were considered. The same reaction temperatures and times as those reported in each of the

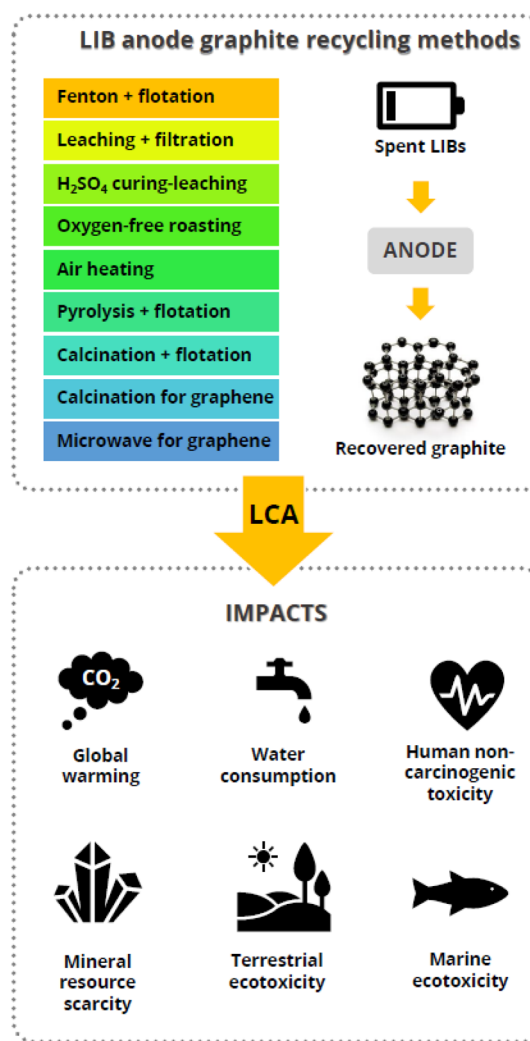


Figure 1. Selected nine lithium-ion battery anode recycling processes and the most representative impact categories for the conducted life cycle assessment analysis.

studied processes (based on published manuscripts) were used. To make this study applicable into a large geographical area, high-voltage electricity from the European Network of Transmission System Operators (ENTSO-E) was applied, which represents 42 electricity transmission system operators from 35 countries across Europe. Further specifications regarding the instruments used for each process are given in the Supporting Information.

Different graphite recycling/recovery processes involving hydrometallurgical and pyrometallurgical processes are here studied, from which the description is given hereafter. For instance, He et al. applied a Fenton reagent-assisted flotation process involving H₂O₂/Fe²⁺ to recycle spent anode and cathode materials from LIBs (process 1, denoted as Fenton + flotation).³⁷ The organic outer layer covering anode and cathode materials was removed during the Fenton reaction so that both the cathode and anode can be simply separated by flotation taking advantage of the hydrophilic (polar) character of the LiCoO₂ cathode and the hydrophobic (nonpolar) nature of graphite. As opposed to other works where low-quality graphite was separated by filtration as a filter, Ma et al. developed a scalable hydrometallurgical recycling process to obtain a high-quality graphite anode (process 2, denoted as leaching + filtration).³⁸ The sorting step was avoided, and recycled graphite was obtained from the filtration residue. Alternatively, low-temperature sulfuric acid curing–leaching together with high-temperature calcination can be applied to recycle and subsequently regenerate spent graphite (process 3, denoted as H₂SO₄ curing–leaching).²¹ In the first step, sulfuric acid was used as

Table 1. Material and Energy Input Inventory of Recycling/Regeneration Processes of the Nine LIB Anodes for a Standardized 100 kg Graphite Treatment^a

Fenton + flotation		leaching + filtration		H ₂ SO ₄ curing-leaching		oxygen-free roasting		air heating	
material	quantity	material	quantity	material	quantity	material	quantity	material	quantity
DI water	7500	DI water	495.33	DI water	250	DI water	15621.8	DI water	447.151
H ₂ O ₂	25.3	H ₂ SO ₄	4.67	H ₂ SO ₄	1015	N ₂	52.14	H ₂ O ₂	0.893
dodecane	2.26	NaOH powder	100	liquid argon	30.68 L			H ₂ SO ₄	8.934
FeSO ₄								N ₂	37.52
methyl isobutyl carbinol	1.13							phenolic resin	7.314
electricity	7150.74 kWh	electricity	76.62 kWh	electricity	252.37 kWh	electricity	956.8 kWh	ethanol	117.066
water	7565.62	water	373.68	water	200	water	15621.8	water	447.151
CO ₂	35.99	H ₂ SO ₄	100	H ₂ SO ₄	1065	graphite	6.892	H ₂ O ₂	0.893
Al	7.6448	Li	0.8119	Li	0.051	Li ₂ CO ₃	0.719	H ₂ SO ₄	8.934
Co	89.467	Al	0.6965	Al	0.1309	N ₂	52.14	N ₂	37.52
Cu	4.9196	Co	0.7354	Co	0.2750	Co	4.587	ethanol	117.066
		Cu	0.2802	Cu	0.0589				
		Ni	0.1088	Ni	0.0148				
		Fe	0.0039	Fe	0.0909				
Mn	2.5865	Mn	2.898	Mn	0.0448				
		Na	0.0611	F	0.1789				
pyrolysis + flotation		calcination + leaching		calcination for graphene		microwave for graphene			
material	quantity	material	quantity	material	quantity	material	quantity	material	quantity
DI water	2200 L	DI water	914.82	DI water	55268.2	DI water	6451.86	DI water	6451.86
N ₂	8.318	HCl	52.42	H ₂ SO ₄	2358.50	H ₂ SO ₄	2113.65	H ₂ SO ₄	2113.65
dodecane	26.4	Ar	2.14	HCl	1.68	HCl	9	HCl	9
				NaNO ₃	85.92	H ₂ O ₂	68.16	H ₂ O ₂	68.16
methyl isobutyl carbinol	13.2			H ₂ O ₂	186.87	H ₃ PO ₄	241.26	H ₃ PO ₄	241.26
electricity	102.95 kWh	electricity	91.60 kWh	KMnO ₄	214.28	KMnO ₄	462	KMnO ₄	462
				electricity	189.29 kWh	electricity	202.157 kWh	electricity	202.157 kWh
				C ₆ H ₈ O ₇	128.24				
water	2200 L	water	766.80	water	55268.24	water	6451.86	water	6451.86
N ₂	12.848	Al	0.51	C ₆ H ₈ O ₇	128.24	KMnO ₄	462	KMnO ₄	462
dodecane	26.4	Li	0.47	Li	3.07				
methyl isobutyl carbinol	13.2	Cu	0.77	KMnO ₄	214.28	H ₃ PO ₄	241.206	H ₃ PO ₄	241.206
2-methylpentane	0.3174	HCl	41.93	H ₂ O ₂	186.87	H ₂ O ₂	68.16	H ₂ O ₂	68.16
3-methylpentane	0.6564			NaNO ₃	85.92	HCl	9	HCl	9
hexane	2.7294			HCl	1.34	H ₂ SO ₄	2113.65	H ₂ SO ₄	2113.65
				H ₂ SO ₄	2358.50	Cu	23	Cu	23

^aQuantities are provided in kg if not stated otherwise. ^bThe full output list corresponding to the pyrolysis + flotation process is shown in the Supporting Information.

an oxidant, while graphite and reaction products acted as reducing agents, transforming the high-valence oxides and metals to their corresponding sulfates. The sulfates were then leached out using additional sulfuric acid (where residual Ni, Co, and Mn can be reused), and a final high-temperature calcination was applied to remove organic materials and regenerate graphite.

Regarding pyrometallurgical approaches, an oxygen-free roasting method combined with wet magnetic separation was applied to recycle cobalt, lithium carbonate, and graphite from mixed electrodes (process 4, denoted as oxygen-free roasting).³⁹ Graphite was used as a reducing agent for the LiCoO₂ cathode during smelting. Interestingly, no chemical solution was required, limiting the generation of secondary pollution. In another work, graphite was recovered and regenerated following air heating and subsequent pyrolysis processes (process 5, denoted as air heating).⁴⁰ After the first shear-emulsion step in a H₂SO₄ + H₂O₂ solution, an air heating step at 300–600 °C was applied to remove the additives from the anode. Graphite regeneration was then carried out by coating a layer of pyrolytic carbon through a sintering process at 950 °C under a N₂ atmosphere.

The separation of the cathode and anode materials is recognized as a complex task due to the organic additives present in the batteries. Zhang et al. proposed a solution to efficiently separate graphite from spent LIBs using pyrolysis, avoiding the need for toxic Fe²⁺ ions (which in turn contaminate the anode) or organic solvents such as *N*-methyl-2-pyrrolidone, dimethylformamide, or dimethyl acetamide.²⁴ To that end, a pyrolysis step at 400–600 °C followed by a flotation process was applied (process 6, denoted as pyrolysis + flotation). In another work, a combination of two-stage calcination and leaching was applied to purify recovered graphite from spent LIBs (process 7, denoted as calcination + leaching).²³ In this approach, aluminum current collectors were selectively leached using 1.5 M HCl, while copper was first oxidized to copper oxide at 500 °C under an air atmosphere so that it can be easily leached out.

In addition to these works, some approaches also explored the possibility of upcycling graphite using spent anodes as a platform material. Taking advantage of the inherent defect structure of graphite from the anodes and the presence of copper traces within, graphene oxide–copper composites were synthesized through calcination, oxidation, and ultrasonic exfoliation (process 8, denoted as calcination for graphene).⁴¹ Interestingly, the obtained materials showed promising photocatalytic properties useful for the removal of pollutants from wastewater. Similarly, Yang et al. used an ultrasonic process combined with microwave digestion to synthesize graphene oxide (process 9, denoted as microwave for graphene).⁴² A modified Hummers' method involving vitamin C as a reducing agent was applied to reduce the environmental impacts of the process. During this procedure, layers of carbon atom surface materials were stripped from graphite as a result of the occurring oxidation–reduction reactions.

Life Cycle Interpretation. LCA studies were performed using OpenLCA software and the Ecoinvent 3.7 Dataset. A gate-to-gate perspective was followed with the objective of solely focusing on the recycling/regeneration phase. The material and energy input inventory was estimated for the treatment of 100 kg of graphite, while 1 kg of regenerated graphite was used as a functional unit. This allowed taking into account the different graphite recovery rates for each process, as recovery rates from ~40 to ~95 wt % were obtained depending on the approach. Although the environmental impacts on the battery field are often reported against electrochemical performance (1 kWh of storage capacity as a functional unit),^{32,43,44} we considered the amount of recovered graphite as a functional unit because at this point, it is difficult (and even unreliable given the data available) to perform graphite recycling LCA studies according to the electrochemical performance of the recycled material. Additionally, only four of the nine recycling processes here studied reported electrochemical studies, so normalizing against electrochemical performance would limit the choice of works to be analyzed and thus lead to distorted results.

RESULTS AND DISCUSSION

Global Warming Potential of LIB Anode Recycling.

Midpoint LCA provides a broad understanding on the environmental impacts of graphite recycling processes. We first focus our attention toward the global warming potential (GWP) impact because it serves as a simple yet effective means to obtain a cross-field comparison with previous studies. Results in Figure 2a are reported based on 1 kg of recovered

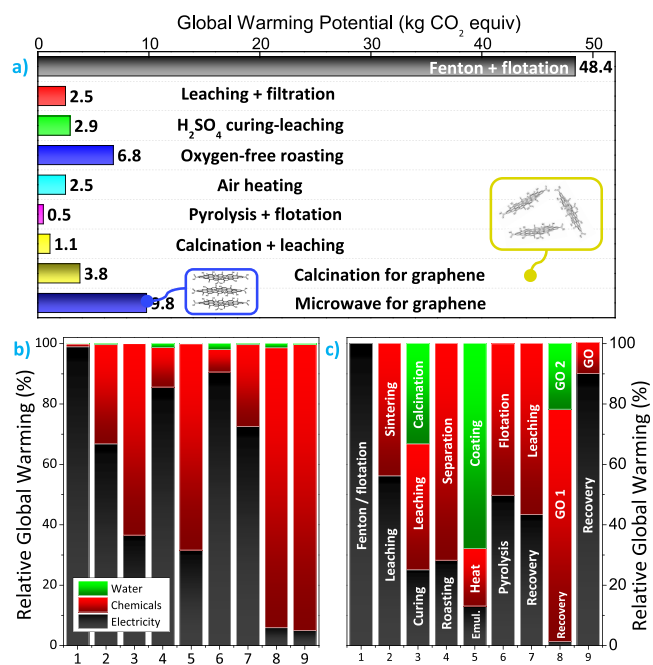


Figure 2. Global warming potential of graphite recycling processes from spent lithium-ion batteries: (a) GWP values in kg-CO₂ equiv. emissions for 1 kg of recycled graphite from spent LIBs. For the process calcination for graphene, the impacts originating from the upcycling of spent graphite to graphene oxide are also represented by a yellow rectangle, yielding a GWP of 42.49 kg-CO₂ equiv.·kg_{graphene-oxide}⁻¹. For the process microwave for graphene, the impacts account for the upcycling of spent graphite into graphene oxide (represented as a blue rectangle). (b) Relative CO₂ contribution of electricity, chemicals, and water used for each graphite recycling process. (c) Relative CO₂ contribution from each step during graphite recycling. Further details on each step are disclosed in the flowcharts provided in the Supporting Information as Figures S1–S9.

graphite from spent lithium-ion batteries. With the exception of the Fenton + flotation process, which yields considerably large values, a GWP ranging from 0.53 to 9.76 kg-CO₂ equiv.·kg_{graphite}⁻¹ is obtained, leaving no doubt that graphite reclamation approaches are environmentally competitive with virgin graphite production (GWP in the range of 1 to 5 kg-CO₂ equiv.·kg_{graphite}⁻¹ will be commented later). Thanks to process simplicity, energy efficiency, and high recovery rates, hydro-metallurgical processes show improved GWP values of 2.49 and 2.89 kg-CO₂ equiv.·kg_{graphite}⁻¹ for leaching + filtration and H₂SO₄ curing–leaching, respectively.^{38,21} The large GWP of 48.41 kg-CO₂ equiv.·kg_{graphite}⁻¹ for the Fenton + flotation³⁷ process can be explained by the low recovery rate of 56.38 wt %, the large liquid:solid ratio of 75:1, and the multicomponent character of the process using 0.1 M FeSO₄, 0.1 M H₂O₂, methyl isobutyl carbinol, *n*-dodecane, and large amounts of energy. Moreover, several assumptions become necessary to complete the inventory modeling due to the lack of accurate

Table 2. Environmental Impacts for 1 kg of Recycled Graphite From Spent LIB Anodes

impact category	recycling process										microwave for graphene	units
	Fenton + flotation	leaching + filtration	H ₂ SO ₄ curing-leaching	oxygen-free roasting	air heating	pyrolysis + flotation	calcination + leaching	calcination for graphene				
fine particulate matter formation	0.07438	0.00565	0.02963	0.01144	0.00348	0.00087	0.00165	0.00725	0.05775			$\times 10^{-3}$ kg PM _{2.5} equiv.
fossil resource scarcity	13.86192	0.69235	1.82762	1.93770	1.79605	0.15196	0.30001	1.15295	4.26566			kWh equiv.
freshwater ecotoxicity	1.64303	0.13917	0.32547	0.22288	0.10028	0.01790	0.0676	0.18210	1.21888			kg of 1,4-DCB
freshwater eutrophication	0.05246	0.00160	0.00248	0.00684	0.00168	0.00055	0.00148	0.00187	0.0068			$\times 10^{-3}$ kg P equiv.
global warming potential	48.40820	2.49542	2.89288	6.82342	2.56400	0.53297	1.08565	3.83233	9.76486			kg CO ₂ equiv.
human carcinogenic toxicity	262.69895	15.44915	27.39227	35.23889	12.93236	2.97395	8.44870	20.9796	208.51649			kg 1,4-DCB
human noncarcinogenic toxicity	12561.18926	630.74599	1828.94960	1651.76153	465.90813	132.86992	398.33412	778.26238	5111.43542			kg 1,4-DCB
ionizing radiation	35.93417	0.73664	1.56213	4.50663	0.70169	0.36732	0.41187	0.95068	1.92872			$\times 10^{-1}$ kBq Co-60 equiv.
land use	1.28048	0.06357	0.10490	0.17030	0.04695	0.01364	0.02709	0.93793	0.90631			$\times 10^{-2}$ m ² a crop equiv.
marine ecotoxicity	15468.24527	798.64296	2241.66621	2041.16128	586.85706	164.00793	494.67243	982.93531	6759.88208			$\times 10^{-1}$ kg 1,4-DCB
marine eutrophication	0.00377	0.00015	0.00017	0.00049	8.41×10^{-5}	3.95026×10^{-5}	0.00011	0.00220	0.00035			$\times 10^{-4}$ kg N equiv.
mineral resource scarcity	10.50620	0.02701	0.04352	0.00808	0.00757	0.00068	0.00159	0.02022	0.36158			$\times 10^{-2}$ kg Cu equiv.
ozone formation, human health	0.09140	0.00664	0.00761	0.01289	0.00618	0.04188	0.00228	0.00901	0.03059			$\times 10^{-2}$ kg NO _x equiv.
ozone formation, terrestrial ecosystems	0.09525	0.00671	0.00784	0.01302	0.00689	0.06677	0.00230	0.00918	0.03124			$\times 10^{-2}$ kg NO _x equiv.
stratospheric ozone depletion	3.64807×10^{-5}	2.97169×10^{-6}	2.08103×10^{-6}	5.04×10^{-6}	1.21×10^{-6}	4.00796×10^{-7}	7.26757×10^{-7}	2.35979×10^{-5}	6.07168×10^{-6}			$\times 10^{-6}$ kg CFC11 equiv.
terrestrial acidification	0.18054	0.01038	0.09735	0.02571	0.00925	0.00203	0.00464	0.02546	0.17343			$\times 10^{-2}$ kg SO ₂ equiv.
terrestrial ecotoxicity	36.53333	4.46955	42.05026	5.34771	2.59456	0.54666	1.19181	7.56926	80.66487			kg of 1,4-DCB
water consumption	1.07585	0.07676	0.29126	0.36751	0.04769	0.03871	0.04717	0.20707	0.64639			$\times 10$ m ³

data. Therefore, it may be reasonable to think that these results overestimate actual GWP impacts.

Pyrometallurgical processes are useful to recycle spent graphite. The air heating process bears a low GWP of 2.56 kg-CO₂ equiv·kg_{graphite}⁻¹ because of the efficient use of H₂SO₄ (only 8.9 kg), H₂O₂ (0.9 kg), and electricity (phenolic resin-ethanol solution is used to regenerate graphite).⁴⁰ Remarkably, pyrolysis + flotation and calcination + leaching processes show GWP values as low as 0.53 and 1.08 kg-CO₂ equiv·kg_{graphite}⁻¹, respectively. Those results are ascribed to the process simplicity, solely requiring nitrogen, argon, water, dodecane, methyl isobutyl carbinol, or HCl, and the high recovery rates of 80.8 and 95 wt %, respectively.²³ The oxygen-free roasting pyrometallurgical process treats both the anode and the cathode, so an additional flotation step (requiring further energy and large amounts of liquid) is applied to isolate graphite, increasing the GWP up to 6.82 kg-CO₂ equiv·kg_{graphite}⁻¹. Finally, the processes aimed at graphite upcycling show acceptable CO₂ emission values regarding graphite recovery, with GWP values of 3.83 and 9.77 kg-CO₂ equiv·kg_{graphite}⁻¹ for calcination for graphene and microwave for graphene, respectively. Those results are interesting considering that the latter process yields a graphite oxide powder instead of regenerated graphite, where oxygen functionalities are introduced to expand the interlayer separation to ~0.71 nm from the ~0.34 nm characteristic of graphite and make the material hydrophilic. From this point, graphene oxide could be obtained following a relatively simple chemical reduction approach. Actually, through the implementation of additional steps in the calcination for graphene process (see Figure S8 for the detailed process flowchart), it is possible to obtain graphene oxide with a GWP of 42.49 kg-CO₂ equiv·kg_{graphene-oxide}⁻¹. The production of graphene oxide from spent batteries brings a 10-fold increase in the CO₂ emissions when compared with the graphite material as graphically depicted by the yellow rectangle with the graphene oxide structure (see Table S11 for the impact distribution for the 18 categories). However, these values are in the low range of those reported by Zhang et al. for graphene oxide production by chemical oxidation at the laboratory scale, with values ranging from 46 to 280 kg-CO₂ equiv·kg_{graphene-oxide}⁻¹.⁴¹

To put these results into context, we should bring our attention to the environmental impacts of raw graphite, either synthetic or natural. Despite the large weight fraction that represents graphite within the batteries, there is a significant lack of data available regarding the environmental impacts of either synthetic or natural graphite production. Synthetic graphite is obtained from raw materials (pet coke and coal tar pitch) having their own embodied environmental impacts. Additionally, large energy is consumed during the high-temperature graphitization step, yielding considerable NO_x, SO_x, and PM₁₀ emissions due to sulfur, nitrogen, and ash impurities in the coal tar pitch and pet coke.⁴⁵ On the contrary, natural graphite has impacts during the mining phase and subsequent purification (requiring large quantities of chemicals such as hydrochloric acid, chlorine gas, or sodium fluoride) up to >99.5% with minimum metal impurities.⁴⁶ With this in mind, the Argonne National Laboratory Greenhouse Gases, Regulated Emissions, and Energy Use in Transportation (GREET) 2018 reported a GWP of 4.9 kg-CO₂ equiv·kg_{graphite}⁻¹ for graphite.⁴⁵ However, the production energy demand was underestimated. In fact, the energy demand was based on secondary industry data and extracted from

thermodynamic estimations assuming that graphite is produced from pet coke and coal tar pitch by carbonation followed by graphitization, although the particle refinement phase needed to obtain battery-grade graphite was not taken into account.⁴⁷ The choice of inert gas during the graphite synthesis process has also been neglected,⁴⁸ potentially leading to a significant environmental impact underestimation. Other studies reported GWP values of 1 to 2 kg-CO₂ equiv·kg_{graphite}⁻¹ (Ecoinvent 3.1) or 5.56 kg-CO₂ equiv·kg_{graphite}⁻¹ for natural graphite.⁴⁹ In any case, Figure 2a shows that some of the processes here analyzed are environmentally competitive with raw graphite and at the same time avoid undesired waste management issues related to uncontrolled battery disposal. These results bring to light the potential of recycling and upcycling approaches of obtain materials with lower environmental impacts.

To evaluate more accurately the environmental impact of each process, the relative distribution to the CO₂ emissions of the electricity/chemical/water mix and each specific procedure (such as leaching, sintering, calcinations, and coating) for each battery is summarized in Figure 2b,c, respectively. Electricity is the main contributor in five of the seven processes recovering spent graphite (average contribution of 69%), while the chemicals have an average contribution of 94% when graphite is upcycled into graphene oxide. Two strategies are envisaged to reduce the global warming effect of graphite recycling. From one side, the scaling of graphite recycling up to industrial volumes may lower the energy consumption, so the overall CO₂ contribution is expected to be reduced in the future. From the other side, transitioning from fossil fuel-based energy to renewable energy generated from natural resources is also encouraged. On the contrary, environmentally friendlier graphite upscaling processes require further emphasis on atom economy and green chemistry principles so that the use of starting materials during the reactions is optimized.⁵⁰ Furthermore, Figure 2c provides an overview of the impact distribution for each subprocess, offering further information for optimization.

Environmental Impacts of LIB Anode Recycling According to 18 Midpoint Indicators. The gate-to-gate environmental impacts of graphite recycling/upcycling per 1 kg of the recovered material are displayed in Table 2. Results are grouped into 18 impact indicators extracted from the life cycle impact assessment. Notable changes are observed depending on the impact category, highlighting the pivotal role of the pyrometallurgical, hydrometallurgical, and combined recycling conditions on the final environmental outcome of the process. These differences could be ascribed to the use of notably different amounts of energy or acids during the recycling process.

To provide a holistic vision, LIB anode recycling processes here analyzed are grouped into hydrometallurgical and pyrometallurgical processes. Generally, hydrometallurgical processes involve spending and generating large volumes of acid solutions, yielding large environmental burdens. For example, HCl is a strong acid causing serious health issues, while it corrodes equipment and yields large amounts of toxic wastewater. Additionally, the metallic impurities extracted from spent graphite or current collector traces (Li, Co, Mn, Ni, or Cu) remain dissolved within the acid/alkali medium, contributing to heavy-metal pollution.⁵¹ The generated large amount of liquid wastes and the produced sludge should be carefully disposed to avoid secondary pollution, and the slow

leaching kinetics under certain conditions (up to 30 h)²¹ increases the economic and environmental cost of this process.⁵¹ However, some of the processes here analyzed are optimized to lower the impacts. In this sense, H₂SO₄ curing–leaching is designed to avoid large energy consumption and generate a relatively low amount of waste (1015 L of H₂SO₄), which coupled with a high recovery rate of 90 wt % yields low impact values. Similarly, the leaching + filtration process solely relies on water, H₂SO₄, NaOH, and electricity (76.62 kWh for the whole process) as it uses room-temperature leaching followed by a quick sintering process with NaOH (500 °C for 40 min).

On the contrary, pyrometallurgical processes use large amounts of inert gases such as argon, contributing to eutrophication, ozone depletion, human carcinogenic toxicity, human noncarcinogenic toxicity, and ecotoxicity.⁵² Pyrolysis processes are also a significant impact driver not only given their energy-intensive character but also due to the release of volatile organic compounds (VOCs) from thermal decomposition, resulting in aerosol hydrocarbon formation with small average particle diameters and potential respiratory risks.⁵³ For instance, graphite readily oxidizes to form CO₂ at temperatures above 700 °C under an oxygen atmosphere. The residual electrolyte and organic binders present in graphite decompose and subsequently volatilize to form harmful gases such as hydrogen fluoride, volatile compounds including ethylene carbonate, propylene carbonate, or diethyl carbonate, and their decomposed products. In this sense, alkali adsorption methods followed by activated carbon adsorption can be applied to avoid the release of these byproducts to the environment.⁵⁴ The calcination + leaching solely requires argon, water, and HCl and a high recovery rate of nearly 95 wt %. The environmental burdens of the oxygen-free roasting process are high as it requires 15,622 L of water, and the process generates 4.6 kg of Co and 0.72 kg of Li₂CO₃ wastes. The high recovery rate of 80.8 wt % and process simplicity of pyrolysis + flotation make this approach efficient environmentally speaking, highlighting the need to consider many aspects when designing environmentally friendly recycling processes.

Interestingly, the microwave for graphene process uses an efficient ultrasound-induced cavitation to separate copper foil and other additives from graphite, reaching dissolution rates with water of 99.5% as opposed with the 57.4% obtained for NMP (despite the nonsolubility of PVDF in water).⁴² However, the use of strong highly toxic chemicals, including H₂SO₄ (2113 kg), KMnO₄ (462 kg), H₃PO₄ (241 kg), H₂O₂ (68.2 kg), and HCl (68 kg), results in large impact indicators. Those results are in line with the report by Arvidsson et al., who quantified the cradle-to-gate impacts of graphene production.⁵⁵ Notable contributions in the ecotoxicity and human toxicity categories were found for the Hummers process. In any case, obtained graphene can replace CRMs such as beryllium (for conductive applications), cobalt (for battery cathodes), gallium (for semiconductors), germanium (for optical fibers), indium (for transparent electrodes), tantalum (for capacitors), or tungsten (for superalloys).^{15,56} In any case, special attention should be paid to the possible environmental and health effects.⁵⁷

The current battery industry is facing a bottleneck regarding access to primary raw materials. In this sense, the “mineral resource scarcity” category merits particular attention because it relates to the use of key raw materials.⁵⁸ As shown in Table 2, this category ranges from a minimum of 0.00068 × 10⁻² kg

Cu equiv. for the pyrolysis + flotation process to 10.50620 × 10⁻² kg Cu equiv. for the Fenton + flotation process. When looking into more detail and considering average values (graphite upcycling methods were also kept out), processes involving hydrometallurgical methods score nearly 7 times more than the pyrometallurgical methods in the mineral resource scarcity category (up to a 2300 increase considering the Fenton + flotation process). These results are explained by the fact that hydrometallurgical processes require abundant inorganic acids, while pyrometallurgy generally avoids the need of such reagents.

Despite the amount of waste generated by hydrometallurgical processes, the results in Table 2 show that this method is environmentally preferred over pyrometallurgical processes. Those results correlate well with the recent work of Mohr et al., who found that advanced hydrometallurgical recycling, in this case for LIB cathodes, shows the highest environmental benefit.⁵⁹ Importantly, our studies consider a 0% recovery of solvents as opposed to the practical solvent recovery rates of 0 to 90% found in industry in the form of recirculation and reuse of the reaction solution.⁵⁵ In this sense, an acid-solvent recovery during the treatment of carbonaceous materials has the potential to lower the water and toxicity footprints. Although the impacts are standardized based on 1 kg of recovered graphite, the maximum material recovery is not *per se* translated into an improved environmental performance.⁵⁹ We should also bear in mind that most of the analyzed processes are designed to improve the purification of graphite (physical separation of graphite from other compounds). In other words, the processes are aimed to yield a high-quality graphite material with impurities in the range of 10 to 100 mg·kg⁻¹. To that end, large quantities of acids are used, so generated water-soluble compounds can be easily removed through rinsing with water (hydrometallurgical method).⁴⁶ Alternatively, high-temperature methods are also used to generate a gas phase of condensed complexes so that impurities with low boiling points could be removed (pyrometallurgical method).⁴⁶ Besides relying on hazardous chemical syntheses (the use of strongly oxidizing inorganic acids or the formation of toxic gases such as hydrofluoric acid during high-temperature treatment), the purification optimization is obtained at the expense of resource/energy efficiency and process simplicity, so little attention is paid to green chemistry principles.⁵⁰ A possible alternative to lessen the environmental impacts associated with anode recycling is the development of one-pot batch syntheses using either low quantities of acids (hydrometallurgy) or shortened heating steps (pyrometallurgy). However, it should be considered that recovering Li from spent graphite does not only alleviate its shortage but it also reduces the ~31.03 mg·g⁻¹ Li content in spent anodes,⁶⁰ exceeding the desired levels from the human and environmental safety point of view.

Sensitivity Analysis. As shown in the materials and energy inputs in Table 1, some of the approaches rely on the use of extremely large amounts of inorganic acids, making the process not only environmentally but also economically nonviable. The excessive use of inorganic acids can also seriously threaten human, animal, and plant life,²⁵ so limiting their use should be a priority. Green chemistry principles also encourage the reduction of hazardous chemical syntheses and the design of synthetic methods for maximizing incorporation of all materials used during the process into the final product, that is, atom economy.⁵⁰ For example, up to 1015 L of H₂SO₄ is

required in the H₂SO₄ curing–leaching method. Based on the assumption that an efficient anode recycling could be achieved by lowering the amount of acids, a sensitivity analysis has been carried out to quantify how reducing the amount of the used acid is translated into improved environmental indicators. The H₂SO₄ curing–leaching method was selected to conduct a sensitivity analysis as it can be considered as a representative approach using industrially available technology in the battery recycling field.²¹ Moreover, the data used for this inventory was verified by the original authors, while several assumptions need to be made in other recycling methods due to lack of available information. To conduct our sensitivity analysis, a graphite:sulfuric acid ratio of 4:1 was applied instead of the original 1:1. As lower amounts of H₂SO₄ are required, the use of deionized water and electricity is lowered from the original 250 to 213.25 kg and from 252.37 to 247.85 kWh, respectively (smaller furnaces with lower energy consumption could be used), while the same amount of liquid argon is computed (the high-temperature calcination step remains unchanged). As no relevant changes on the impurity removal during the curing process with H₂SO₄ are observed in the study, no changes on waste flows were implemented.

The effect of the H₂SO₄ reduction on 18 impact indicators in a gate-to-gate LCA perspective is summarized in Figure 3.

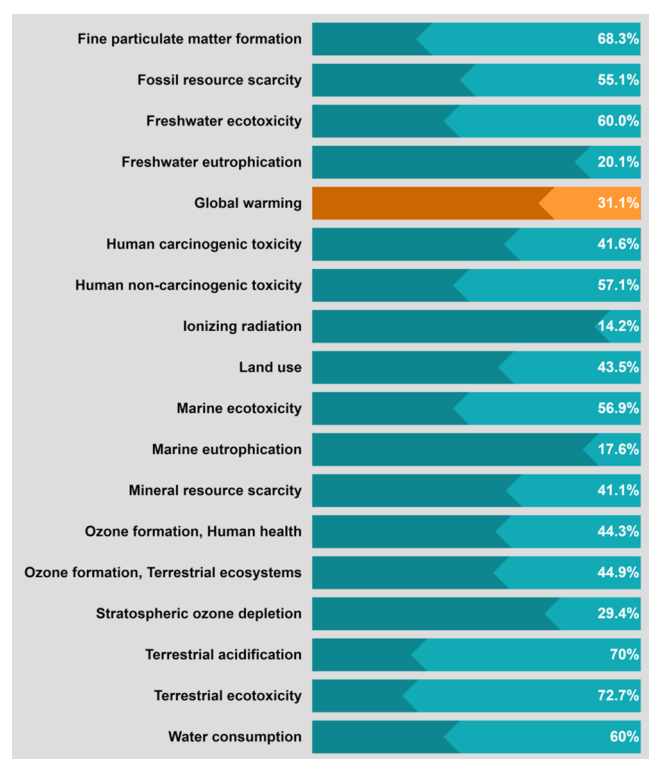


Figure 3. Sensitivity analysis of graphite recycling based on a modified H₂SO₄ curing–leaching method with a graphite:sulfuric acid ratio of 4:1 instead of the original 1:1.

For the sake of clarity, relative reduction on each impact indicator is provided in the vertical axis. The environmental impacts are reduced by 14.2 to 72.7% depending on the category. In eight of the 18 impacts, the reductions exceed 50%, with special improvements in categories related to toxicity (terrestrial acidification and ecotoxicity, fine particulate matter formation, freshwater ecotoxicity, marine ecotoxicity,

and human noncarcinogenic toxicity). With a pK_a value of 1.92 at 25 °C, sulfuric acid is classified by the international agency for research on cancer as a group I carcinogen.²⁵ Additionally, sulfuric acid has a high toxicity, with LC₅₀ values (lethal concentration 50) of 1.67 and 2.9 mg·L⁻¹ for fish and aquatic invertebrates after 48 h.²⁵ Regarding the GWP, a reduction of 31.1% achieved (to 1.99 kg·CO₂ equiv.·kg_{graphite}⁻¹) makes this procedure more competitive against virgin battery-grade graphite (either synthetic or natural). So, at this point, the following question arises: is it worth using so many toxic components to obtain marginal improvements in the graphite quality and its electrochemical performance? In this sense, further works need to be carried out to study the quality of recovered graphite under modified parameters to optimize the process so that it can be ready for scale-up.

Electrochemical Performance. As the energy storage field is a performance-driven science, the electrochemical performance of regenerated graphite needs to be also discussed to provide an improved understanding on the applicability of the actual different recycling/regeneration processes. Accordingly, Table 3 summarizes the electrochemical performance of recycled/regenerated graphite in a Li/graphite half-cell configuration (theoretical capacity of 372 mA h·g⁻¹). Additional details regarding anode fabrication and the electrochemical performance of commercial graphite are also provided to enable comparison.⁶ To prepare the battery, regenerated graphite is usually mixed with a polymeric binder and a conductive filler in a solvent-based process, and the obtained slurry is coated onto metallic foil to act as a working electrode (copper). Upon battery charging, Li⁺ is released from the cathode material, and after crossing through the electrolyte, it is intercalated into the graphite layers at the anode to form an intercalation compound (i.e., LiC₆). A poor electrochemical performance of recycled graphite is related to a combination of a large impurity content²¹ with a damaged graphite structure hindering Li⁺ intercalation.⁶¹

As depicted in Table 3, regenerated graphite from the leaching + filtration process yields graphite delivering 377 mA h·g⁻¹ at 0.1C and retaining 84.6% of its initial capacity after 100 cycles at 0.2C (304 mA h·g⁻¹).³⁸ Such a high capacity was ascribed to the well-defined crystalline hexagonal phase (space group: *P63/mmc*) and to the fact that added C65 (conductive carbon black additive) may contribute to the capacity. However, the delivered capacity rapidly decays to 120 mA h·g⁻¹ at 1C due to a large charge transfer resistance (*R*_{ct}) of 105 Ω, which hinders the migration of Li⁺. Regenerated graphite from H₂SO₄ curing–leaching offers an initial capacity of 349 mA h·g⁻¹ at 0.1C, while it retains 98.9% of its initial capacity after 50 cycles at 0.1C.²¹ This result, comparable to that of commercial graphite (351 mA h·g⁻¹ after 50 cycles at 0.1C), is ascribed to the appropriate carbon atom layer spacing and the high purity (99.6%) of regenerated graphite and a low *R*_{ct} of 37 Ω. Despite the better rate performance in comparison with the previous process (200 mA h·g⁻¹ at 1C vs 120 mA h·g⁻¹), commercial graphite behaves better at high rates (260 mA h·g⁻¹ at 1C), suggesting that the structure of regenerated graphite was not fully recovered during recycling. The structural defects in graphite resulting in the formation of a thick solid electrolyte interphase (SEI) were found as a primary reason for the lowered storage capacity at high rates.²¹

Among the pyrometallurgical processes, air heating yields graphite with a high tap density of 1.03 g·cm⁻³ as opposed to 0.90–0.95 g·cm⁻³ of commercial battery-grade graphite

Table 3. Electrochemical Performance of Recycled/Regenerated Graphite in a Li/Graphite Half-Cell Configuration^a

recycling process	binder	additive	solvent	electrolyte	delivered capacity (mA h·g ⁻¹)	cycling stability	rate capacity (mA h·g ⁻¹)	R _{ct} (Ω)
Fenton + flotation ³⁷	N.R.							
leaching + filtration ³⁸	10 wt % PVDF	10 wt % C65	NMP	N.R.	~377 at 0.1C (the added C65 may contribute to the capacity)	84.6% after 100 cycles at 0.2C	~270 at 0.5C ~120 at 1C ~30 at 2C	105.0
H ₂ SO ₄ curing–leaching ²¹	5 wt % PVDF	3 wt % AB	NMP	1 M LiPF ₆ in EC:DMC:EMC	349 at 0.1C	98.9% after 50 cycles at 0.1C	~282 at 0.5C ~200 at 1C ~45 at 2C	37.0
oxygen-free roasting ³⁹	N.R.							
air heating ⁴⁰	4 wt % PVDF	1.5 wt % AB	NMP	1 M LiPF ₆ in EC:DEC	377 at 0.1C	98.8% after 50 cycles at 0.1C ^b	~320 at 0.5C ~285 at 1C ~265 at 2C	N.R.
pyrolysis + flotation ²⁴	N.R.							
calcination + leaching ²³	5 wt % PVDF	5 wt % AB	N.R.	1 M LiPF ₆ in EC:DMC	591 at 0.1C (graphite interlayer spacing and pore size increase)	97.9% after 100 cycles at 1C	~320 at 0.5C ~180 at 1C	~305
calcination for graphene ⁴¹	upcycling: improved photocatalytic activity against methylene blue, ~92% after 300 min vs ~34% for bare CuO							
microwave for graphene ⁴²	upcycling: ordered graphene with a resistivity of 41.2 Ω·m							
commercial graphite ²¹	5 wt % PVDF	3 wt % AB	NMP	1 M LiPF ₆ in EC:DMC:EMC	352 at 0.1C	99.7% after 50 cycles at 0.1C	~322 at 0.5C ~260 at 1C ~103 at 2C	31.4

^aR_{ct}, charge transfer resistance obtained from Nyquist plots. PVDF, polyvinylidene fluoride; C65, a type of carbon black; AB, acetylene black; NMP, *N*-methyl pyrrolidone; EC, ethylene carbonate; DMC, dimethyl carbonate; EMC, ethyl methyl carbonate; DEC, diethyl carbonate. 1C = 372 mA·g⁻¹. Cycle capacity and rate capacity values are extracted from plots by the naked eyes, so an error of ±5 mA h·g⁻¹ is possible. N.R., not reported. ^bOnly reported for the charge cycle.

(interesting for larger volumetric capacities).⁴⁰ In this line, regenerated graphite offered a capacity of 377 mA h·g⁻¹ at 0.1C, keeping 98.8% of its initial capacity after being charged/discharged for 50 cycles at 0.1C. When increasing the cycling rate to 0.5C, 1C, and 2C, remarkable capacities up to ~320, 285, and 265 mA h·g⁻¹ were obtained. Those results are explained in terms of a large specific surface area of 11.47 m²·g⁻¹ obtained upon regeneration, offering an intimate contact with Li⁺ and enabling a fast Li⁺ intercalation/deintercalation. More interestingly, graphite recycled following the calcination + leaching method allowed a remarkable initial discharge capacity at 0.1C of 591 mA h·g⁻¹.²³ This large capacity (372 mA h·g⁻¹ theoretical capacity of graphite) is explained by a thinner and smaller particle size with a larger interlayer spacing and pore size (Ø of 13.5 nm), which stores additional Li at the graphite edges and surfaces. Good cycling stability was also obtained as denoted by the 99.7% retention after 50 cycles at 0.1C. Despite the high capacity achieved at low cycling rates, when the rate increases to 0.5C and 1C, the delivered capacity decays to 320 and 180 mA h·g⁻¹. Those results can be explained by a large R_{ct} of nearly 305 Ω, which impedes a fast Li⁺ intercalation/deintercalation. Overall, these results show that regenerated graphite could efficiently work at cycling rates up to 0.5–1C, although the damaged structure cannot compete with the performance delivered by virgin graphite at high rates.

Although not applied as energy storage materials, the upcycling of graphite into graphene-based nanocomposites deserves special mention. In this context, the calcination for graphene process transforms graphite from spent LIBs into graphite oxide, graphene, and finally graphene oxide–copper composites.⁴¹ Thanks to its large specific surface area and

oxygen groups, which promote the formation of free radicals, the obtained material showed an improved photocatalytic decomposition of methylene blue under UV light, reaching degradation levels of ~92% after 300 min in comparison with ~34% achieved for bare CuO. Despite not being studied for a specific application, graphene resulting from the process microwave for graphene showed a resistivity (Ø = 10 mm sample) of 41.2 Ω·m,⁴² which can be beneficial for different energy storage and electronics applications. Upcycling is a bright approach to valorize waste materials as feedstock resources for value-added applications while it minimizes waste accumulation in the environment with alternative fate with respect to landfills.^{62,63}

Perspective into the Future. New efforts are being directed to avoid secondary pollution originating from strong acids during hydrothermal treatments. In this framework, Markey et al. used 5 wt % boric acid as the leaching reagent to upcycle graphite anodes from spent LIBs.⁶⁴ Boric acid, a nonvolatile and noncaustic acid, presents significant environmental advantages over the acids commonly used for graphite reclamation. For instance, boric acid shows an NFPA 704 rating of 1-0-0 (health-flammability-instability) in comparison with the 3-0-2 of sulfuric acid or the 3-0-1 of hydrochloric acid. Followed by a sintering process, the residual Li trapped in the bulk of graphite particles is completely removed, and besides healing the damaged graphitic structure, this method creates functional boron doping onto graphite surfaces. As a result, obtained graphite delivers 332 mA h·g⁻¹ at C/3 and retains ~100% of its initial capacity after 100 cycles (C/3).

Based on the results here obtained, we estimate that both hydrometallurgical and pyrometallurgical approaches have room for improvement regarding their environmental perform-

ance when it comes to spent graphite recycling. Hydrometallurgical methods should optimize the amount of acids used to limit toxicity and mineral resource scarcity issues, while reducing reaction times and temperatures would lower energy consumption. Pyrometallurgy uses inert gases and generally requires additional energy input, so using renewable energy and improved reaction times/temperatures should be pursued. At this point, a combination of hydrometallurgy and pyrometallurgy (as the Umicore battery recycling process does) seems to be environmentally preferable because it scores notably better in relevant impact categories including global warming, freshwater toxicity, human toxicity (carcinogenic and noncarcinogenic), marine ecotoxicity, mineral resource scarcity, terrestrial acidification, terrestrial ecotoxicity, and water consumption.

Many positive technological signs for the battery field are flourishing within the “refurbish, reuse, and recycle” Circular Economy perspective. Paired with the legislative efforts for the implementation of electric vehicles and their reuse/recycling, a bright future moving away from manufacturing technologies relying on virgin materials is yet to come. In this sense, the European Commission has set a minimum recycling rate of 60 wt % for LIBs (battery pack without housing; oxygen and hydrogen excluded) by 2030.⁶⁵ In this framework, this study aims to provide orientation toward practical decision making for the design of environmentally friendlier battery recycling alternatives.

Raw materials (extraction, transport, and purification) hold a considerable share of the carbon footprint (and other environmental impacts) of batteries. For example, graphite accounts for up to 40% of the greenhouse gases, 63% of SO_x emissions, and 67% of NO_x emissions regarding the cradle-to-gate impacts of 1 kWh LiNi_{1/3}Mn_{1/3}Co_{1/3}O₂-type LIBs.⁶⁶ Taking into account that only in Europe, approximately 270,000 tons of battery-grade graphite will be required to reach the goal of 300 GWh·year⁻¹ of battery production (European Battery Alliance),³³ novel approaches for the environmentally friendly production of graphite are urgently needed. The substitution of petroleum-based feedstock for synthetic graphite production can be a partial solution. However, new holistic alternatives are required to obtain battery-grade graphite with a greater environmental sustainability. In line with previous works highlighting the potential of recycling to significantly improve the environmental performance of LIBs, vanadium redox-flow batteries, or aqueous hybrid ion batteries over their life cycle,^{67,59} graphite recovery from spent LIBs could lower the environmental footprint during battery production and concomitantly solve the end-of-life scenario. Upon the gradual incorporation of recycled anodes into the existing primary processing facilities, the harmful effects of anode waste on human health and the environment would be reduced.

Several graphite recycling approaches fail to analyze how obtained graphite can be applied again into an LIB, leaving aside a relevant aspect when considering the recyclability of materials, specifically the reuse in real conditions. We estimate that recycling approaches need to also focus on electrochemical measurements to provide an extended life cycle approach. In addition, to enable the practical implementation of graphite recycling processes, the relatively poor rate performance should be addressed. Taking into account that several battery chemistries share the same electrochemical principle (ion intercalation/deintercalation), graphite recycled

from exhausted LIBs can be also applied in environmentally friendlier options such as potassium-ion batteries.⁶⁸ Finally, to ensure the industrial feasibility of laboratory-scale studies, the use of existing metallurgical infrastructure should be preferred.

CONCLUSIONS

With the aim of enabling the practical design of industrially scalable and environmentally friendly lithium-ion battery recycling processes, the environmental impacts of spent graphite recycling and regeneration processes are quantified. Life cycle assessment is applied to obtain the overall environmental profile of pilot-scale (100 kg) recycling processes with a gate-to-gate perspective. Energy and material inputs are disclosed to facilitate future follow-up studies. Hydrometallurgical and pyrometallurgical recycling processes, together with graphite upcycling methods, are analyzed, and environmental impacts are assembled into 18 standardized ReCiPe 2016 midpoint impact categories. Global warming values from 0.53 to 9.76 kg·CO₂ equiv. per kg of recovered graphite, freshwater ecotoxicity values of 0.015–1.64 kg of 1,4-DCB, human carcinogenic toxicity values ranging from 3 to 263 kg of 1,4-DCB, and mineral resource scarcity values varying from 0.00068 to 10.5 × 10⁻² kg Cu equiv. are achieved. As current graphite recycling processes remain mostly focused on obtaining highly purified graphite with maximum recovery rates, large quantities of inorganic acids are used to leach out residual Li, Co, Mn, or Cu. In fact, the specific impacts can be reduced by up to 73% by solely limiting the amount of used acids. The intensive use of primary energy is the other main underlying factor behind observed large impact values. These results clearly highlight the need of implementing LCA during the early design process so that the environmental hot spots over the entire process can be recognized, bringing us closer to a true circular battery industry with reduced environmental impacts. At this stage, combined processes comprising hydrometallurgical and pyrometallurgical steps give environmentally preferable results as they score notably better in the most relevant impact categories.

Given the versatility of carbonaceous structures, there is a plethora of opportunities to develop increasingly more sustainable variants to classical graphite, particularly considering a cradle-to-grave life cycle perspective. For example, activated carbons or carbon aerogels, which function well as anodes in LIBs, could be obtained using naturally available abundant precursors that do not compete with the food chain.⁶⁹ Coupled with the development of less energetically intensive synthetic processes, considerable reductions on CO₂ equiv. emissions and toxic elements may be achieved. While the most environmentally sustainable LIB anode procedures are selected and these processes are scaled up, scientists and industry could be also inspired on the rational design of biomass-based carbons. Merging the advances in both directions can bring us one step closer to more sustainable energy storage systems.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acssuschemeng.1c04938>.

Flowcharts and tables summarizing the inventory modeling and environmental impacts and a brief

flowchart description for each recycling process to guide the reader (PDF)

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Author Contributions

The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript.

Notes

The authors declare no competing financial interest. Table ¹ provides the full material and energy input inventory. The Supporting Information shows the summarized flowcharts of each of the analyzed processes, together with a brief explanation to guide the reader and the inventory modeling for materials and energy implemented from the Ecoinvent 3.7. database. The calculations performed to obtain the full inventory are available from the corresponding author upon reasonable request.

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