



Review Article

Multi-source recovered graphite and its use in electrodes for energy storage

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Abstract

This review summarises the latest improvements in the recovery of graphite to be used in electrochemical energy storage (EES) devices, without limiting to lithium-ion batteries as the only source and final destination of graphite. The work is focused on the characteristics of graphite recycled by different processes and its effect on the electrochemical behaviour of the application. An analysis of the advantages and disadvantages of each method was carried out considering factors related to the environment, cost, and usefulness of the recovered material. Finally, some challenges and our perspectives are outlined.

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Keywords

Recycling, Anodes, Electrodes, Carbon, Batteries.

Introduction

The production of waste is increasing every day and, if humankind does not act responsibly, the degradation of the environment can become irreversible. The rule of the three Rs (Reduce, Reuse & Recycle), which is a proposal for consumption habits, aims to develop

conduct that makes the planet a more sustainable place. This seems to be a practice that can provide answers to current problems related to pollution and global warming [1].

The use of practically everything can be reduced, but beyond that, there are a lot of materials (or even devices) whose reuse is somewhat limited, mostly due to their own intrinsic characteristics. In these cases, the most important factor in preventing them from becoming waste is recycling. For example, this is the case of Lithium Batteries (LIBs) used in cars and in portable devices. Their main reuse is fundamentally limited to static energy storage where energy density is not a crucial factor (houses and offices, with no major problems regarding volume and weight) [2]. On the other hand, recycling them, or rather recycling their components, is growing day by day and is expected to increase in the coming decades, mainly because of regulations that are motivating them to do so [3,4].

Regarding that, due to the price and the scarcity of materials such as cobalt, the first efforts to recycle LIBs components were focused mainly on their cathodes [5]. Today, due to a variety of factors (legal, economic, geopolitical, and practical) work is beginning to be done on anode recycling in order to recover graphite, a much less expensive material than that contained in the cathode but worthy of being recovered [6]. It is important to bear in mind that a commercial LIB anode has a graphite content of more than 90% [7], which increases the need for selective, effective, and efficient recycling.

Currently, graphite used in the production of anodes for Li batteries is obtained from two sources: natural and synthetic [8]. The growth in demand for the production of energy storage devices has led to the consumption of natural resources, and these will be exhausted if no action is taken to recover them [9]. On the other hand, the use of synthetic graphite has a considerable carbon footprint because the process requires large amounts of energy [10].

This way of acting is not only convenient in the sense of not accumulating waste (with the environmental and health issues that this generates), but it also reinforces

the *circular economy* [11] of the material or the product. This means that regions that do not contain the natural resources for their production can also have access to these raw materials. This is known as *urban mining* [12], that is, the extraction of an element that is now found in high concentrations in a city deposit as a result of the accumulation of end-of-life devices [13].

LIBs were discussed in the first instance due to the magnitude of their production and growth [14], but the recovery of graphite can be done in several industries with very diverse by-products and waste.

For example, in the production of aluminium, in addition to the use of carbon cathodes (which need to be changed every 5–8 years and can be recycled, 10 kg/t of Al) [15], during the aluminium smelting process, there is what is known as “pot lining” which is produced at a rate of 22–25 kg/t of aluminium generated [6]. One specific kind of this pot lining used is constructed from the carbonaceous cathodes utilized in the electrolysis of alumina, and consequently, this is a rich source of graphite.

In this context, various methods were developed to recover graphite [16]. Each of them has its advantages and weaknesses. Likewise, the quantity and quality of recovered material can be very different. At first, simple characterization and calculations are enough, but then, in view of its new use, in addition to a complete morphological, compositional, and structural characterization, it is necessary to assemble and test the electrochemical devices, at least on a laboratory scale. Typically, but not exclusively [17], LIBs are the destination for most of the graphite production.

Here, in this mini-review article, we will focus on recently developed methods for recycling graphite obtained from various sources and its use in electrochemical energy storage (EES) systems. Special emphasis is placed on the quality of the material obtained as well as on the electrochemical performance achieved.

Discussion

As mentioned above, there are several sources of graphite for recycling. To date, both in terms of the industries currently involved in its recycling and regarding the number of scientific papers published, the main source is still LIBs anodes, which produce the largest volume. It is worth mentioning that LIBs are also the main target for the recycled graphite obtained, both in industry and in academic reports. The state-of-the-art is analysed below based on the latest articles reported in the scientific literature, and it is summarised in [Table 1](#).

The electrochemical performance of recycled graphite from both pyrometallurgy and hydrometallurgy (or a

combination of both) is highly dependent on the physical and chemical processes employed. The search for protocols to obtain graphite with extremely low contaminant loadings (Cu, Al, Li, Fe, Si, Mn, F, P, etc.), better known as “battery grade graphite” (>99.95% C), has led to the use of recovery methods and the application of purification treatments that are particularly aggressive to the carbon structure of the graphite as well as being highly expensive and polluting. This, together with the characteristic wear suffered by the material in its first life as an anode, will be decisive in defining the aptitude of the graphite and its potential second life in a given electrochemical device.

This has been discussed in a series of recent papers proposing the recovery of graphite by a combination of thermal and aqueous treatment of the materials of interest. The electrochemical response of the graphitic anodes obtained is further affected by the efficiency of the protocols aimed at the purification of metal contaminants, the removal of binders, and restructuring the carbon chain in consecutive steps. Using this strategy, M Bhar. et al. recovered graphite from discarded 18,650 type cylindrical LIBs (used in laptops) by three consecutive treatments [18]: **washing** in water, then in dimethyl carbonate (DMC), and finally **heating** at 750 °C in Ar in order to remove non-conductive organic oligomers, lithium salts and other impurities. In this work, the different electrochemical responses of the materials were evaluated, with special emphasis on the effect of the application or not of the second step (DMC washing). In this sense, after a careful formation of the solid-electrolyte interphase (SEI), cycling at different C-rates was performed, and in long term essays (1000 cycles), there were excellent results at 0.5C. The report of so many cycles is remarkable, as this is not very common in the scientific literature of the field. Graphite without the DMC wash showed an efficiency of 91.8%, and with the wash it was 93.12%, considering the initial capacities of 305 and 318 mA h g⁻¹, respectively. As well as retaining a specific capacity close to that of pristine graphite (372 mA h g⁻¹), the good performance of the anodes could also be evidenced in the capacity retention after the rate capability study, whose evolution occurred with a Coulombic efficiency of 99.9% in a fully reversible process. Finally, the potential of the recycled materials when tested in a full-cell configuration is noteworthy (*vs.* NCA-811 cathodes), although only 30 cycles were reported. On the other hand, it is worth highlighting the potential of the recovery protocol proposed by Chenxing Yi and co-workers [19]. By means of an industrially scalable thermal process, they obtained graphite anodes with high specific capacity (360.8 mA h g⁻¹), great capacity retention (99.8%), and good cycling stability at 1C. The promising electrochemical responses in half-cells make this recycled graphite a potential candidate for direct application systems. For this reason, it would have been interesting to know its performance in the

Table 1

State of art of Graphite Recovery from electrochemical devices and others sources.

Ref	Recycle		Extraction procedure		Purity		Electrochemical Performance				
	From	To	Step 1	Step 2	Carbon (%)	Impurities	Specific Capacity (mA h g ⁻¹)	C – rate/ Specific current	Cycle number	Capacity Retention (%)	Full Cell Application
[19]	LIB	LIB	Heat treatment 1400 °C–4 h	Ultrasonication in H ₂ O 15 min - sieving	99.5	Cu = 0,435% - Li < 0,01% - P = 0,03%	360.8	1.00C	100	99.8	–
[34]	LIB	LIB	Mechanical - Acid leaching	600 °C 1 h H ₂ SO ₄ 2 M–12 h Co(NO ₃) ₂ –900 °C 4 h	–	Li, Al, Mn, Co, Ni, Cu < 50 mg/kg Fe 171.9 ± 5.2 mg/ kg	245.8	1.00C	500	–	–
[18]	LIB	LIB	DI water leaching	750 °C–5 h - Ar.	–	Li, P, F, O based. Low level.	305	0.50C	1000	91.8	–
				DMC wash- 750 °C–5 h - Ar.	–	–	318	0.50C	1000	93.1	NCA-811 -30 cycles - 0.35 mA h, 4.3–3 V at 30 mA g ⁻¹
[20]	LIB	LIB	(NH ₄) ₂ S ₂ O ₈ leaching	Vacuum drying	98.76	Cu 1.3 ppm Al, Li, Fe < 0.2 ppm	365.3	0.10C	500	–	LiFePO ₄ - 50 cycles - 0.5C – 90 mA h g ⁻¹
							330.2	1.00C	500	–	
[25]	LIB	LIB	Electrolysis (electrode as cathode) 30 V - 2.00 M Na ₂ SO ₄ – 25 min	–	94.5–95.6	F based 4,31–5,12% Na <0.023% Ca, Cu, Fe, Si, Al < 0.01%	373.9	0.50C	100	97.54	–
[35]	LIB	LIB	Citric Acid leaching	–	–	Cu, Na, Al < 0.1 ppm	330	0.50C	80	–	–
[26]	SC – AP	LIB	NaOH - Ethanol washing 450 °C–2 h	HCl washing Vacuum drying	92.6	Aluminosilicate, SiO ₂ ,	357.6 ± 9.6	0.1C	50	90.4	–
[27]	SC – AP	LIB	NaOH 1 M–3 h	H ₂ SO ₄ 0.5 M–3 h	88.27	N = 0.19%, S = 0.32%	280	0.10C	100	–	–
[28]	LIB ^a , Alk, Zn/C–B	LIB	^a 2.00 M H ₂ SO ₄ /H ₂ O ₂ (5% v/v)	^a Low temperature drying	^a 73.65	^a O = 14.86%, Si = 1.88%, Al = 0.81%, Cu = 0.80%, F = 7.85%, P = 0,045%	370	0.50C	20	–	–
[36]	Zn/C–B	LIB	Ultra-pure water Electrolysis 30 V–150 h	–	–	Fe = 3.32% Si = 2.74% Al = 1.66% Ca = 0.49% K = 0.47% Ti = 0.32% Ni = 853 (ppm) Zn = 603 (ppm) Mg = 362 (ppm)	458	0.20C	150	96	–
[30]	LIB		Manually disassembly		85.58		–	–	–	–	

(continued on next page)

Table 1. (continued)

Ref	Recycle		Extraction procedure		Purity		Electrochemical Performance				
	From	To	Step 1	Step 2	Carbon (%)	Impurities	Specific Capacity (mA h g ⁻¹)	C - rate/ Specific current	Cycle number	Capacity Retention (%)	Full Cell Application
		Non faradaic capacitor		Hummers reaction Thermal reduction 400 °C 30 min		O = 8.14% Al = 6.28%					Capacitor -Capacitance 156 F g ⁻¹ at – 5 mV s ⁻¹
[31]	LIB	NIB	Manually disassembly	Ethanol wash 1300 °C–4 h – Ar.	91.91	O = 8.09%	162	0.20 A g ⁻¹	1000 (2.00 A g ⁻¹)	94.6%	–
		KIB					320	0.05 A g ⁻¹	200 (0.02 A g ⁻¹)	74%	–
[32]	LIB	LSB	Manually disassembly	NMP wash, precipitation and water wash	85.32	–	–	–	–	–	LSB - 500 cycles. - 0.5C –765 mA h g ⁻¹
[33]	LIB	ACDIB	Manually disassembly	Sonication – water washing 650 °C–5 h – Ar.	–	–	250	372 mA g ⁻¹	300	–	ACDIB - 50 cycles - 4.4 V –58 mA h g ⁻¹ - Ret: 70%

long term (i.e., 1000 cycles) as well as in a full-cell configuration.

It is evident that there is a relationship between the electrochemical performance of recycled graphite anodes and the equitable removal of contaminants (inorganic and organic) by hydrometallurgical treatments when the latter are combined with thermal treatments. The good electrochemical characteristics, such as cycling capacity and stability, indirectly indicate that the recycled materials have stable SEI and an sp² structure that is able to react during the electrochemical discharge process. However, the future of graphite anode recycling should turn to options where the use of processes, such as thermal treatment, with high energy demand and effluent emissions is dispensed with, i.e., moving rapidly towards the implementation of purely hydrometallurgical processes.

In this sense, the development of processes that include the use of reagents and solvents that can be fed back into the same protocol or, as a last resort, be reused in other production processes is of great importance for battery recycling. The use of metal complexing agents, chemical oxidants, and modifications in the pH and temperature of the reaction bed are some recently proposed alternatives. In one such process proposed by Hougui Xiao and co-workers, graphite was recovered by separating it from the current collector and binder in a one-step process using **ammonium persulphate** and **water** [20]. The authors claim that simultaneous separation of the active material from the anode composite is possible, leaving behind fluorinated impurities from the binder and metallic impurities from the current collector. In addition, the process promotes the de-intercalation of Li from the graphene structure. It remains in solution to be recovered later by selective precipitation. The characterisation of half-cells with recycled graphite anodes by this route resulted in a high reversible capacity of 365.3 mA h g⁻¹ after 100 cycles at 0.1C and 330.2 mA h g⁻¹ after 500 cycles at 1C. On the other hand, the use of the anodes in full-cells for 50 cycles demonstrated an acceptable electrochemical performance, showing a reversible capacity of 95.6 mA h g⁻¹ with 72.8% retention (*vs.* LiFePO₄ cathodes, theoretical capacity value:170 mA h g⁻¹).

Another interesting hydrometallurgical alternative is recycling through the use of membrane electrochemical reactors. The advances made in both the extraction of Li from brines and the recovery [21] of discarded cathode materials [22] are presented as an environmentally sustainable alternative. Two reasons reinforce this hypothesis. The first is that the effluents produced as a consequence of the electrochemical process are minimised because they can be recirculated in the process or reused in other battery recycling stages, or the reagents involved can be recovered for commercial purposes

(acids and bases). The second reason is that energy from natural sources is considered as a source of power [23]. Through solar and wind energy, electrolytic processes with appropriate experimental design can contribute to reducing the carbon footprint of the recycling process. Particularly where water splitting is the driving force, obtaining hydrogen as a by-product promises a further reduction in energy consumption because it could subsequently be used to power fuel cells [24]. For example, graphite recycling is possible by aqueous **electrolysis of the anodes in saline solution**. The principle is similar to that discussed above. In an electrolytic cell, the anode obtained from discarded batteries is placed in front of a graphite plate, as the positive electrode, and immersed in 2 M Na₂SO₄ aqueous solution, which only acts as a supporting electrolyte. A potential difference of 30 V is applied between them for 25 min. On one hand, the hydrogen formed at the negative electrode promotes the separation of the discarded anode components. On the other hand, it facilitates solubilisation of the remaining Li together with other metal contaminants. As a result, with this method, N. Cao and co-workers obtained a copper current collector completely free of graphite, which was electrochemically characterised [25]. At different C-rates, the graphite half-cells showed somewhat irregular profiles, possibly due to the presence of impurities (Fluorine based 4.31–5.12%). Although the reported electrochemistry presents good values, mainly in the cycles performed between 0.1 and 1C, anomalous profiles are remarkable both in the rate capability experiment (capacity increase with cycling) and in the cycles at higher C-rates (2C and 5C). The best electrochemical performance was obtained at a 0.5C, where a capacity retention of 97.54% was achieved after 100 cycles, considering an initial value of 409.5 mA h g⁻¹. A capacity recovery of 373.9 mA h g⁻¹ was also achieved after cycling at different high C-rates.

In the continuous search for more efficient, economical, and less polluting methods, advances in the state of the art of dismantling, extracting, and recovering battery components has driven the development of innovative technologies that arise from a **combination of the classical hydro and pyrometallurgical** processes. This is no exception in the case of graphite, where the possibility of recycling from other sources has also been assessed.

As previously mentioned, spent graphite electrodes from the **aluminium** industry (SC-AP) are among other possible sources for obtaining graphite for recycling. In this regard, the group of Tian, Peng *et al.* made use of the **alkali fusion method** (NaOH, 450 °C for 2 h and HCl washing), and after removing 31% of impurities, they obtained graphite with a purity of 92.6% [26]. By several characterisation techniques, they reported that both the degree of crystallisation and interlayer spacing of the recovered graphite were unevenly distributed. In addition, the existence of many defects and even

graphene in the material were observed. Unlike commercial graphite that needs to be activated prior to its use as an anode, the graphite obtained by this method, containing amorphous carbon and slightly expanded graphite, could skip this step. Nevertheless, they achieved good results using it in half-cells (267.8 ± 12.5) mA h g⁻¹ after 310 cycles at 1C. A very important point to highlight in this work is the statistics they performed, as they tested more than one battery for each experimental value and reported accordingly.

Another recently reported method for the recovery of electrodes from the **aluminium** industry is based on two-stage **caustic/acidic leaching** [27]. The final result was graphite with 88.27% purity. The residual percentage is constituted by several non-electroactive compounds (identified by XRD) which then decrease the performance when used as an anode in LIBs. In this regard, the authors demonstrated a considerable improvement in battery cycling both in capacity and stability compared to simple water washing and even caustic leaching methods. Both improvements are related to the removal of impurities. The first one is the elimination of non-electroactive material, and the second improvement results from an increase in the stability of the SEI. Thus, the best result obtained, without using any additives, was 280 mA h g⁻¹ after 100 cycles at 0.1C (or 325 mA h g⁻¹ with the mass corrected for the content of active graphite being a non-conventional way of reporting the result obtained).

It is worth mentioning that when the material to be recycled was not obtained from battery anodes but from larger materials before being used as anode material, it must be ground to the desired particle size, adding an extra and unavoidable step to the process. On the other hand, when the material is ground but agglomerated on a current collector, an extra stage is also required, but it usually requires less energy.

Finally, it is important to highlight obtaining of graphite from primary batteries (alkaline). The work published by Alcaraz [28] and Sawangphruk [29] exemplifies this. In the first case, they used acid leaching (6 M HCl/H₂O₂ (25% v/v)) to obtain graphite from spent **alkaline and Zn–C batteries** and, although they carried out a complete characterisation of the materials obtained from different sources, they only reported electrochemical results for one material, and it was not the one coming from the alkaline batteries that are of interest here. In the second article, the starting point was expired Panasonic neo-D cells. The carbon rods were manually extracted and washed and finally 30 V was applied for 150 h in ultrapure Milli-Q water to obtain the material. Here, the values obtained are really impressive (about 458 mA h g⁻¹ at 0.2C for 150 cycles) because they exceed the theoretical values expected for lithium

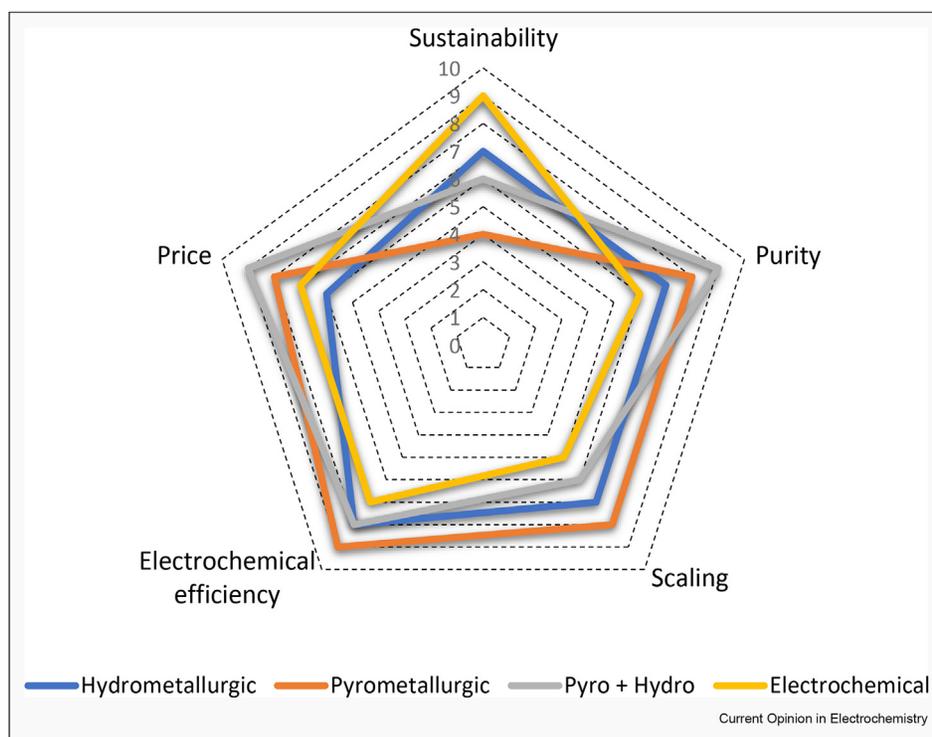
intercalation in LiC_6 (372 mA h g^{-1}), and the precise explanation given by the authors was the existence of extra capacity coming from the insertion of Li in LiC_3 structures, which was facilitated by the presence of defective, graphene-like, multi-layered, carbon nano-sheets. They mainly support this with electrochemical evidence: plateaus at 1.2 and 2.5 V in the galvanostatic charge.

At this point, it is important to note that while most recycled graphite is used in LIB anodes, this is not the only EES system in which it has been studied. In this line, Mohapatra et al. reported the use of GO and rGO synthesised by a modified Hummer's method, avoiding any curing treatment, and they used spent LIBs as the starting material to fabricate **non-Faradaic supercapacitor** electrodes [30]. The rGO synthesised by this method showed better electrochemistry than rGO obtained by the thermal route and commercial rGO. The authors attribute this to the improved interconnection of the structure and the absence of restacking of the graphene sheets. The best specific capacitance reported was 156 F g^{-1} in a neutral electrolyte. It is worth mentioning that to treat 1 g of graphite, the authors used 50 mL H_2SO_4 (98%), 4 g KMnO_4 , 50 mL of distilled water, 5 mL of H_2O_2 , and HCl until neutralised.

In addition, a study by Wu and co-workers demonstrated, for the first time, the use of recycled graphite in both **sodium-ion batteries** (NIB) and **potassium-ion batteries** (KIB) [31]. The graphite was obtained by manual extraction from the anodes of spent LIBs, followed by ethanolic washing, and finally calcining at different temperatures between 700 and 1600 °C for 4 h in Ar. After an exhaustive morphological, compositional, and electrochemical characterisation of the material (including *in-operando* XRD), the authors reported the following values: in the case of the NIB, 162 mA h g^{-1} at 0.2 A g^{-1} with 80% of capacity retention at 5 A g^{-1} and 94.6% after 1000 cycles at 2 A g^{-1} ; in the case of KIB they reported capacities of 320 mA h g^{-1} at 0.05 A g^{-1} with a 74% of capacity retention at 0.02 A g^{-1} after 200 cycles.

Regarding new battery technologies, the group of Yang and co-workers was one of the pioneers in the use of recycled graphite in **lithium-sulphur batteries** (LSB) [32]. To do this, they used spent LIBs from Mercedes Benz cars, from which they extracted graphite from the anode by dissolving the binder with NMP followed by precipitation and washing with water. The cathode was prepared by infiltrating a high sulphur content into the carbon (78.4%) but a low sulphur mass loading on the electrode (1 mg S cm^{-2}). Finally, using

Scheme 1



Schematic representation of the characteristics of the methods used in graphite recycling.

CR2025 cells and conventional electrolytes for this technology (1 M LiTFSI in DME/DOL (v/v = 1:1) + 1 wt % LiNO₃), they achieve very good cyclability values. These were even better than those obtained with the use of commercial graphite. According to the authors, this was due to improvements in pore structure (from 0.0073 cm³ g⁻¹ to 0.0143 cm³ g⁻¹), additional defects, polar functional groups, and the presence of transition metals. It is remarkable that the maximum temperature used in the treatment was 70 °C.

A somewhat particular proposal can be found in the article published by Santhanagopalan and co-workers [33]. There, the authors used graphite from commercial LIBs (3.2 V, 5 A h, cylindrical 32,650 cell) after being cycled 1000 times at 1C. The method chosen for its recovery was based on sonication, water washing, and heat treatment (three temperatures were tested). The recovered graphite was used in a Li-ion based **all-carbon dual-ion battery** (ACDIB). Thus, with the graphite obtained by heat treatment at 650 °C, the anode half-cell provided a specific capacity of 250 mA h g⁻¹ at 372 mA g⁻¹ with a cycling stability of 300 cycles (PF₆⁻ intercalation). On the other hand, with the graphite obtained by the 1000 °C heat treatment, the cathode half-cell showed a first discharge capacity of 65 mA h g⁻¹ at 100 mA g⁻¹ with a stability of 100 cycles (Li⁺ intercalation). Finally, the full-cell configuration delivered an initial discharge capacity of 58 mA h g⁻¹ with an average discharge voltage of 4.4 V, and it performed 50 cycles with a capacity retention close to 60%.

Considering the large difference in production costs between cathode materials and graphite, we must consider whether recovery and reuse in new batteries is really viable. In other words, it must be supported with the greatest possible scientific rigour whether bringing graphite to battery-grade purity really means a process of environmental pollution or, on the contrary, only contributes to increasing the carbon footprint associated with the production of energy storage devices. This is a topic that has been addressed in the literature, and various alternatives have been proposed for its second use in other energy storage devices such as capacitors and also for the synthesis of graphene materials, adsorbents, etc.

Conclusions and perspectives

According to this review of the latest advances in graphite recovery for use in EES, the origin of the material and the recycling processes influence the subsequent electrochemical performance of the assembled cells. This is mainly related to impurities that have not been removed and depends on the structure that has developed in the material obtained.

An in-depth and comparative analysis of this work can be graphically summarised in [Scheme 1](#). The five key properties of this analysis have been detected and evaluated, producing a schematic diagram that helps to visualise a comprehensive comparison.

Although the vast majority of the experiments carried out only used graphite in half-cell devices, these developments showed great potential for laboratory-scale applications. Recovered graphite showed more than acceptable electrochemical performance; in many cases, it was better than commercial graphite cells. All this must take into account that there are no standard experiments. This is why each research group assembles the electrodes and cells and reports the data in its own way. This makes a rigorous comparison impossible, both for the electrochemical report and for the constitution of the electrode itself, and even for the type of cell used.

Furthermore, the methods presented effectively reduce the cost of graphite as an input, although it is not possible to state the same for the cost of treating the effluents produced in its recovery.

In most cases, advances have focused on the replacement of polluting solvents with low or zero effluent production processes using reusable aqueous solutions (acidic, basic, or neutral saline) as well as reducing the temperatures used or even eliminating the steps where heat is used. It is worth highlighting the emerging methods which use electrochemical processes for the recovery of anode materials and obtain the required electric energy from renewable sources. For these reasons, it is necessary to develop industrially scalable and sustainable recycling technologies that make it possible to produce high quality materials from the large volumes of waste expected in the near future.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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