Recycling is a potential solution to narrow the gap between the supply and demand of raw materials for lithium-ion batteries (LIBs). However, the efficient separation of the active components and their recovery from battery waste remains a challenge. This paper evaluates the influence of three potential routes for the liberation of LIB components (namely mechanical, thermomechanical, and electrohydraulic fragmentation) on the recovery of lithium metal oxides (LMOs) and spheroidized graphite particles using froth flotation. The products of the three liberation routes were characterized using SEM-based automated image analysis. It was found that the mechanical process enabled the delamination of active materials from the foils, which remained intact at coarser sizes along with the casing and separator. However, binder preservation hinders active material liberation, as indicated by their aggregation. The electrohydraulic fragmentation route resulted in liberated active materials with a minor impact on morphology. The coarse fractions thus produced consist of the electrode foils, casing, and separator. Notwithstanding, it has the disadvantage of forming heterogeneous agglomerates containing liberated active particles. This was attributed to the dissolution of the anode binder and its rehardening after drying, capturing previously liberated particles. Finally, the thermomechanical process showed a preferential liberation of individual anode active particles and thus was considered the preferred upstream route for flotation. However, the thermal treatment oxidized Al foils, rendering them brittle and resulting in their distribution in all size fractions. Among the three, the thermomechanical black mass showed the highest flotation selectivity due to the removal of the binder, resulting in a product recovery of 94.4% graphite in the overflow and 89.4% LMOs in the underflow product.

KEYWORDS: lithium-ion battery, automated mineralogy, liberation, froth flotation, recycling

1. INTRODUCTION

Lithium-ion batteries (LIBs) are widely used nowadays in various devices, from portable electronics to electric vehicles. Growing global demands for Co, Mn, Ni, Li, and graphite, which are present in LIBs, have further stressed the already scarce supply of such raw materials worldwide. These surging demands can lead to supply risks, price fluctuations, and market monopoly. Effective recycling strategies are thus necessary to reduce the need of virgin raw materials and to promote resource preservation and environmental protection. Such efforts are exemplified in the current stipulation by the European Union, where a minimum recycling efficiency of 50 wt % for LIB (Directive 2006/66/EC) is required, with an ongoing proposal to reach 70 wt % by 2030. Furthermore, the European Union categorized Li as a critical raw material in 2020, while Co and natural graphite have been considered as such since 2010, highlighting their economic importance and supply risks.

The current and emerging recycling technologies for LIBs typically focus on the recovery of components that have high economic value, such as Co, Ni, and Cu. To reach a higher recycling efficiency, new products such as graphite should be considered, as it represents 15–20% of LIB’s total mass. More robust recycling strategies are sorely needed. It is also noteworthy that despite the aforementioned
regulations, the supply risk of graphite has only recently gained attention. Indeed, graphite will remain an essential component of LIBs in the foreseeable future. According to a survey carried out by Benchmark Mineral Intelligence, anode materials are forecasted to experience the highest increase in demand among all the raw materials used in LIBs, that is, +700% between 2018 and 2028. Currently, China has a dominant position regarding this critical material, accounting for 64% of the global graphite production and almost 90% of anodic spherical graphite. As a result, Western countries are actively searching for strategies to develop their own graphite supply chain for battery production, including plans to integrate recycled graphite.

During typical LIB recycling processes, a comminution stage is used as a first stage to separate electrode particles from their current collectors, producing foils in the coarse size fraction and active particles in the fine fraction. This is possible since electrode foils present a ductile behavior and, as reported by Schubert, ductile materials can be liberated through shear, cutting, and tearing stresses. The resulting fine fraction is typically referred to as “black mass” and contains mixed cathode and anode active materials.

Most of the state-of-the-art recycling technologies use black mass resulting from the pretreatment (e.g., mechanical or thermomechanical pretreatment) as a starting point for chemical processes for metal recovery. Indeed, in industry, this black mass is usually not further sorted and is directly fed to either thermomechanical (TM) or electrohydraulic fragmentation (EHF). The impact of each strategy on the characteristics of black mass components is the presence of a polymeric binder, which is important information in order to understand the electrode behavior during recycling. The nature of the binder, which is important information in order to understand the electrode behavior during recycling, is a major factor for the anode components.

This paper provides insights into the influence of three liberation processes prior to froth flotation: mechanical (M), thermomechanical (TM), and electrohydraulic fragmentation (EHF). The impact of each strategy on the characteristics of black mass particles was studied using automated mineralogy. This novel approach for black mass characterization was recently proposed by Vanderbruggen et al. and Dade et al. and provides information on the chemical composition, morphology, and degree of liberation of LIB components. While most research on LIB recycling has focused on the operating conditions and design of separation processes, there is a need to gain a fundamental understanding of particle behavior and liberation as a result of treatment stages that systematically affect their overall recycling efficiency. Such knowledge could also support the LIB design and production practices.

2. MATERIALS AND METHODS

2.1. Material and Processing. The material for this study consisted of 200 discharged identical LIBs (Samsung INR18650-29E, batch no BRGA29E82105). One battery was manually opened and dismantled to estimate the mass of each major component, as presented in Figure 1. In this particular case, the active materials were scrapped off manually from the foils. After dismantling, the recovered material was placed in an oven at a maximum of 40 °C for safety reasons for 24 h and then the weight of each fraction was measured with a scientific balance (ENTRIS6202I–1S, Sartorius Lab Instruments GmbH & Co, Germany), and the difference in weight corresponds to the estimated electrolyte, which was evaporated (Figure 1). The electrolyte value is quite low compared to the literature, which can testify that the electrolyte is not entirely evaporated at that temperature and some electrolyte compounds such as the conducting salts might have remained.

Complementary experiments have been carried out to identify the nature of the binder, which is important information in order to understand the electrode behavior during recycling. The electrodes of a dismantled INR18650-29E were immersed in tap water for 10 min. As observed in the Supporting Information (Figure S1), all the graphite particles were thus detached from the Cu foils, confirming the hypothesis of a water-soluble binder for the anodic electrode. For the cathodic electrode, the LMOs remained laminated on the Al foil, testifying to the presence of a binder insoluble in water, likely PVDF.

For this study, the batteries underwent three recycling routes (Figure 2), hereby referred to as M, TM, and EHF, to determine the influence of each process on the liberation of battery components and their eventual recovery. The pretreatment processes are described next.

1. The M process used a single-shaft rotary impact shear crushe UG300MS (MeWa universal granulator, Andritz, Schwäbisch, Germany) operated at a peripheral speed of
6.4 m/s with a bottom grid of 10 mm. The feed consisted of intact batteries producing shredded materials passing through the discharge grate, while coarser particles undertake further stress. The operating principle of this crusher is the comminution of the feed material by applying shearing stress between a block-shaped rotor and stator tools, as detailed by Lyon et al. \(^{27}\)

2. In the TM route, vacuum pyrolysis was performed at 500–650 °C. Subsequently, comminution of thermally treated products was carried out using a Retsch SM 2000 cutting mill (Retsch, Inc., Newtown, USA) with a bottom sieve of 10 mm mesh size.

3. The EHF route employed an EHF unit (EHF-400 ImpulsTec GmbH, Germany). The EHF uses three cathodes at 40 kV to produce shock waves, discharging at a rate of 1 to 4 Hz. The process produced two fractions: float and sink, which were dried in an oven at 80–100 °C. Further information on this equipment is found in Ohl et al. \(^{28}\).

The resulting black mass products (<1000 μm) from M, TM, and the sink fraction from the EHF process were classified with a vibratory column sieve (Fritsch Analysette 3, Idar-Oberstein, Germany) using four nominal sizes: 1000, 500, 125, and 63 μm. For each fraction of the black mass, a representative subsample of 1.5 g was taken using a rotary splitter (PT100, Retsch GmbH, Haan, Germany) for analytical investigations. Each black mass type had five samples, one per size fraction and two epoxy blocks for the fraction 500–1000 μm to minimize particle size effect on samples representativeness.

In addition, the fine fraction (<63 μm) was further processed by froth flotation to separate graphite particles from the LMOs. The fraction below 63 μm was taken using a rotary splitter (PT100, Retsch GmbH, Haan, Germany) for analytical investigations. Each black mass type had five samples, one per size fraction and two epoxy blocks for the fraction 500–1000 μm to minimize particle size effect on samples representativeness.

The preparation of epoxy blocks followed the method as originally suggested by Rahfeld and Gutzmer \(^{30}\) to analyze graphite particles and modified in Vanderbruggen et al. \(^{10}\). The particles were embedded in a homogeneous mixture of epoxy resin and iodoform. To minimize errors and bias caused by sedimentation and settling, vertical slices (B-sections) were used, as suggested by Heining et al. \(^{31}\). These slices were rotated 90° and fixed in a new epoxy resin. The sample was then polished and carbon-coated, thereby creating a conductive surface and scanned by SEM.

The mineral liberation analysis (MLA) system hereby used combines a SEM (FEI Quanta 650F) equipped with two EDX spectrometers (Bruker Quantax X-Flash 5030). The image was...
3. RESULTS AND DISCUSSION

3.1. LIB Component Distribution. Each process was compared according to their individual performances starting from the mass distribution and losses, as presented in Figure 4.

The results indicated that losses of material occur for all the processes from 5.8 to 24 wt %. The M process is the least at 5.8 wt %, likely including fine dust and electrolyte. The electrolyte in LIBs consists of a mixture of organic solvents and conducting salts, such as LiPF₆. A significant amount of solvent may evaporate when exposed to the atmosphere; however, some electrolyte compounds, such as the conducting salts, might have remained in the M-BM. Although new methods have been explored to recover the electrolyte, they require controlled atmospheres or the use of supercritical fluids, such as CO₂.

On the other hand, the EHF process reported 11.1 wt % mass losses corresponding to electrolyte removal, dispersion, or dilution of fine particles such as conductive carbon and graphite and partial binder dissolution.

Finally, the TM process presented the highest losses at 24.2 wt %, which can be due to the decomposition and consumption of organic materials during the vacuum pyrolysis stage. As reported in the literature, the separator decomposes first, then polypropylene (155 °C) and polyethylene (135 °C) followed by binder decomposition, for example, PVDF (450 °C) and SBR (248 °C).

In all processes, the particles associated with casing materials are efficiently recovered in the coarse fraction (<1000 μm). As shown in Figure 5, almost no casing particles were identified in the black mass (less than 0.4 wt %). These heavy particles are usually recovered by an established industrial process after mechanical crushing using a densimetric table, such as those practiced by Retriev Technologies, Recupyl Valibat, AkkuSer, and Accurec.

The main difference between the processes results in the distribution of the cell components. The comparatively low mass of the black mass (36.2 wt %) obtained with mechanical treatment shows that the majority of the components are not comminuted and remained coarse, especially the Cu and Al foils. Furthermore, due to its elasticity, the separator particles remained in the coarse fraction, which can be easily recovered using, for example, an air classifier.
processes implemented derived from automated mineralogy data.

In the case of the EHF process, the separator particles may also be recovered in a relatively simple manner. Indeed, during experiments, these particles were reported to the surface of the aqueous medium due to their low density.

In addition, the distribution of cathode and anode active particles during each processing is distinct. These differences reflect the variable impact that each processing strategy has on the properties of the LIB components. To understand this influence and further evaluate these processes, it is necessary to carry out an in-depth characterization of the resulting material particles.

3.2. Black Mass Characterization. One of the most relevant parameters for the evaluation of the effectiveness of a recycling pretreatment operation is liberation, as it affects the efficiency of downstream separation processes. Therefore, the three black masses (<1000 μm) were further evaluated by automated mineralogy, as shown in Figure 6. The objective of this characterization is to understand the behavior of each species after processing, as will be discussed in subsequent sections.

3.2.1. Mechanical Preprocessing. In the black mass treated via mechanical preprocessing, the active particles, graphite, and LMOs comprise over 95% of the mass in all size fractions (Figure 6). These results are similar to the ones obtained by Widikatmoko et al., where only a few particles of Al and Cu foils were found below 850 μm after milling.

The M-BM contains the least Cu and Al foil residues compared to the other processes, as foils are deformed rather than comminuted in the rotary crusher (Figure 6A). The foils are known to be ductile, non brittle, and have a good tensile strength, requiring more energy and stress for their comminution. The electrode foils were well delaminated from the active particles.

Despite this effective delamination of the active particles from the electrode foils, only 5.1 wt % of black mass was reported to <63 μm (Figure 6B). This is due to the preservation of the polymeric binder, as observed in Figure 7. During the battery production, the active particles are coated with the binder, forming aggregates that are laminated onto the foils using a calendaring process. Thus, mechanical comminution can delaminate but not liberate individual active particles. The aggregates shape resulting from delamination can be observed on the Figure 6A.

3.2.2. Thermomechanical Preprocessing. The volume of fine black mass obtained with the TM route is higher compared to mechanical processing (i.e., 51 wt % than 36 wt %, respectively). In this case, the anode active particles were not evenly distributed within all size fractions, being preferentially concentrated in sizes below 63 μm. Figure 6D shows spheroidized graphite particles that are mostly well liberated, highlighting the full decomposition of the anode binder. Indeed, a binder such as SBR degrades above 248 °C.

Based on several studies, the PVDF cathode binder decomposes above 450 °C and optimal pyrolysis temperatures of 550 °C, according to Zhang et al. Thus, the use of vacuum pyrolysis above 500 °C should eliminate the binders. However, despite the treatment of black mass at 550–650 °C in this work, the cathode electrodes preserved their lamination, as observed in Figure 6C. This preservation could mean that only a partial decomposition of PVDF was achieved. One of the contributing factors for a partial decomposition of the cathode binder might be that entire batteries were fed into the pyrolysis chamber in this study. The compacted structure of the electrodes in the battery may hinder the efficiency of the pyrolysis process because the electrode surfaces are not directly exposed, reducing the decomposition of the binder.

Another explanation might be that Al melted, protecting the laminated structure of the cathode. Also, despite the discharging step for safety, a thermal runaway may have occurred, leading to higher temperatures than expected in some areas of the sample. As seen in Figure 6C, there is a Cu particle containing Al traces with patterns characteristic of melting, suggesting a temperature above its melting point of Al at 660 °C was reached. Al melting is an issue for LMO liberation, as mentioned by Li et al.

The TM-BM contains Al foils in its different size fractions (Figure 6 in the modal composition). Unlike the M-BM, the Al foils after thermal treatment appeared uneven and punctured (Figure 8), probably caused by corrosion promoted by HF in the electrolyte during pyrolysis. The thermal process caused fissures and a subsequent increase in brittleness of the Al fraction. The change in mechanical properties of Al explains its presence in all size fractions of the black mass after comminution. The change in mechanical properties of Al explains its presence in all size fractions of the black mass after comminution. Therefore, it might be recommended to apply the mechanical-thermal (MT) route: first a mechanical process to produce BM with a low amount of fine Al impurity particles to avoid Al melting during the subsequent pyrolysis. Another advantage of conducting the pyrolysis on the BM and not on the input batteries is that it would lead to more uniform binder decomposition due to increase in exposed surfaces, which could facilitate liberation. After pyrolysis, the produced MT-BM should then be milled to liberate the active particles and concentrate them into the fine fraction.

Another aspect of the TM-BM is the difference in metal oxide species present. As observed in Figure 9, only the TM-BM presented a NiCo phase. This suggests that during the thermal process, there was a partial reduction of metal oxides. Yang et al. concluded that Ni ions from NMC were reduced to metallic Ni, while the Co and Mn ions appeared to maintain their trivalent form. Various studies explained that thermal pretreatment can promote the carbothermic reduction of the LMOs and has the advantage of improving their leaching efficiency and decreasing the amount of reducing agents (e.g., H₂O₂) needed during hydrometallurgical treatment.

3.2.3. Electrohydraulic Fragmentation Preprocessing. Among the three routes, the EHF process leads to the highest relative mass of LIB, reporting to the fraction below 1000 μm.
56.3 wt%. The EHF method uses only high voltage pulsations and liberates battery components without comminution tools. After thousands of pulsations, the batteries were completely opened. Most of the active particles were well liberated from the foils and concentrated in the fine fractions of the black mass. The active particles, graphite, and LMOs conserved their spherical shape during this process (Figure 6F). Öhl et al. showed that the LMOs after the EHF process can be reused in new battery.
cell production but at a lower grade than the new material. Alternatively, the EHF-BM can be further refined to pure chemical elements via hydrometallurgy.

The presence of a water-soluble anode binder, such as CMC, SBR, or PAA, helps graphite delamination from the Cu foil and its deaggregation (as shown in the Supporting Information Figure S1). The pH of water after the operation remained neutral (ca. 7). Interestingly, while graphite particles appeared to be well liberated, and its presence in the fraction <63 μm was comparatively lower than for the TM route. A reaggregation mechanism has occurred, leading to heterogeneous aggregates containing liberated active particles, as observed in Figure 6E. This can be due to the anode binder dissolution and its rehardening during the drying stage. Indeed, in LIB production, the water used as a solvent for the anode binder is removed by evaporation before calendering onto the Cu foil.

It is also observed in Figure 10 that most nonliberated cathode active materials remain bound to the Al foil, proving that only the anode binder is dissolving and revealing a different behavior for the anode and cathode binders. Recently, Lyon et al. showed that the combination of mechanical comminution to open the batteries, followed by EHF for decoating of electrodes, presented a higher liberation efficiency for the cathode electrode than the EHF alone.

3.3. Froth Flotation. Froth flotation was used to separate graphite particles from the LMOs based on their difference of wettability. The results of the rougher flotation experiments carried out in this study are summarized in Figure 11. Since the feed of flotation have different grades of graphite and LMOs as a result of the preparation process (Figure 6 modal composition), the flotation efficiencies cannot be directly compared. The M-BM shows a moderate selectivity with a recovery of 63.2% graphite in the froth product and 66.6% LMOs in the underflow product. This comparatively low selectivity is attributed to the residual binder coating active particles, resulting in a decrease in the wettability difference between the graphite and the LMO particles. Indeed, the commonly used cathode binder, that is, PVDF, has a hydrophobic behavior promoting the recovery of LMOs. These results are comparable to the study of Zhang et al., where they floated crushed electrodes and 62.5% of the LMOs were recovered in the underflow product. The low efficiency of graphite recovery may be the result of two conditions that require further study. In the first place, it is difficult to disperse the oily kerosene used as a collector, and thus, it may not be efficiently adsorbed on the graphite surface, even in cases where they are well liberated. Furthermore, if both graphite particles and binder-coated LMOs present similar hydrophobicity, this might result in a competitive flotation effect.

The TM-BM presented the highest selectivity of the samples hereby studied, with a graphite recovery of 94.4% in the froth product and 89.4% LMOs particles in the underflow product. With this treatment process, the active particles present a stronger difference in wettability due to a comparatively superior binder removal. The anode binder is decomposed during the pyrolysis stage, aiding kerosene adsorption on the graphite surface. As mentioned above, the LMO phase has been modified

![Figure 7](image7.png)

Figure 7. SEM image with imposed colors on loose powder of an LMO aggregate from M-BM. The blue spheres are the LMOs, and in between, there is the residual binder.

![Figure 8](image8.png)

Figure 8. Left: SEM image with imposed colors on polished grain mount of a cathode particle. The difference of grey colors shows the Al-oxidation on the side of the foil. Some holes can be observed on the Al foils. The black particles, for instance, on the left of the foil, are the resin which went through the porosity during sample preparation.
during this TM process (Figure 9), which might also affect their wettability, an unexpected phenomenon that should be further studied. With this TM process, the true flotation of the LMOs is diminished due to binder removal. Furthermore, in a recent publication by our research group, it was shown that the flotation efficiency of the TM black mass can be improved by applying an attrition pretreatment. Such pretreatment refreshes the particle surface, removing the anode binder and pyrolysis residues, resulting in an improved graphite flotation. Attrition also removes a part of the cathode residual binder, reducing the true flotation of LMOs. In addition, with this high-shearing pretreatment, there is a disaggregation of graphite and LMOs, decreasing the entrainment of the metals in the graphite overflow product.

The EHF-BM shows graphite recovery in the overflow product of 83.9%. As earlier discussed, it is possible that the anode binder is better dissolved during the EHF process, allowing a better adsorption of kerosene and thus leading to a higher recovery. However, some graphite particles might be trapped in heavy agglomerates and hence not recovered. Nevertheless, the flotation selectivity was harmed by a significant recovery of LMOs in the overflow product (42.2%). Similar to the M-BM, the residual PVDF promoted the recovery of LMO particles, in addition to some possible...
<table>
<thead>
<tr>
<th>Process type</th>
<th>Advantages</th>
<th>Drawbacks</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mechanical (M)</td>
<td>(+) possibility of recovering the electrolyte and separator.</td>
<td>(−) aggregates of active particles under the laminated shape due to the residual binder.</td>
</tr>
<tr>
<td></td>
<td>(+) low content of foils and casing particles in the BM. The BM contains mainly the active particles.</td>
<td>(−) low volume of BM and especially in the fractions below 125 μm due to the conservation of active particle lamination shape.</td>
</tr>
<tr>
<td></td>
<td>(+) good delamination of the foils present in the BM.</td>
<td>(−) residual binder results in a lack of selectivity for flotation. True flotation of LMOs due to coated organic binder.</td>
</tr>
<tr>
<td></td>
<td></td>
<td>(−) mix of active particles in the flotation; therefore, the products have to be sent to hydrometallurgy to recover the metals. The graphite ends up in the leaching cake.</td>
</tr>
<tr>
<td>Thermomechanical (TM)</td>
<td>(+) anode binder well removed, resulting in a high liberation degree of the graphite and copper foils.</td>
<td>(−) electrolyte and separator lost.</td>
</tr>
<tr>
<td></td>
<td>(+) low impurity content in the graphite flotation product, potential for graphite recycling.</td>
<td>(−) Possibility of Al melting decreasing cathode liberation.</td>
</tr>
<tr>
<td></td>
<td>(+) efficient separation of LMOs and graphite with froth flotation due to binder removal.</td>
<td>(−) after pyrolysis, Al foil has a brittle behavior. Al particles are spread in the whole BM. These impurities are challenging for the following hydrometallurgic stage.</td>
</tr>
<tr>
<td></td>
<td>(+) LMO flotation product presents a high content of metals and a really low content of graphite particles compared to the BM. The removal of the graphite particles, which are porous, reduces the acid reagent consumption during hydrometallurgical treatment.</td>
<td>(−) partial reduction of metal oxides during pyrolysis so no possibility of direct recycling of the LMOs. The LMO product needs to go through hydrometallurgical treatment to recover the metals.</td>
</tr>
<tr>
<td></td>
<td>(+) partial reduction of the metal oxides has the advantage to improve their leaching efficiency and decreasing the amount of reducing agents needed during the hydrometallurgical treatment</td>
<td></td>
</tr>
<tr>
<td>Electrohydraulic fragmentation (EHF)</td>
<td>(+) recovery of separator foil (skimming).</td>
<td>(−) electrolyte lost.</td>
</tr>
<tr>
<td></td>
<td>(+) high liberation of the active particles from the foils. A wet screening can further separate the foils from the active particles.</td>
<td>(−) Al and especially Cu impurities, which are not found in the other BM.</td>
</tr>
<tr>
<td></td>
<td>(+) high volume of BM.</td>
<td>(−) if followed by a drying stage, there is rehardening of the soluble binder, leading to heterogeneous agglomerates. Therefore, it is recommended to remain in the wet process.</td>
</tr>
<tr>
<td></td>
<td>(+) liberation of active particles while conserving morphology, so there is possibility of direct recycling of LMOs and graphite.</td>
<td></td>
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</table>
entrapment mechanisms. In addition, the fact that the EHF-BM was dried after the EHF stage might influence the surface, and it should be further studied. Nevertheless, to avoid binder rehardening and, therefore, unselective agglomerates, it is recommended that the BM remains under slurry between the EHF product and the flotation process. These results, compiled together, point out the need to rethink binder chemistry. Indeed, the PVDF binder is one of the biggest challenges during LIB recycling due to its toxicity and the difficulties to remove it, diminishing the cathode component liberation during comminution and also flotation selectivity due to its hydrophobic behavior. Therefore, finding alternatives for the cathode binder chemistry, such as water-soluble binders, which are already commercialized for the anode electrode, would increase the LIB recycling efficiency.

Currently, many different pretreatment methods are established and produce a black mass of different quality. Based on the obtained results, the advantages and drawbacks of each route is summarized in Table 1.

4. CONCLUSIONS

The aim of increasing the recycling efficiency of LIBs requires an understanding of how the LIB components are liberated during the recycling process. This paper characterized black mass samples obtained from different preprocessing routes: M, TM, and EHF. For the characterization, automated mineralogy enabled qualitative and quantitative observations of the properties of LIB components and their dependence on comminution processes. As it is hereby shown, LIB liberation is component-specific, a function of material properties, and influenced by the methods employed, resulting in different black masses. The presence of the binder and its preservation is found to have a strong impact on the separation of active materials using froth flotation.

- With the M process, the active particles were well delaminated from the electrode foils. The BM contained a really low amount of foils and casing impurities. The active particles remained under their lamination shape due to the binder conservation, leading to only a fine fraction of active particles being individually liberated. In addition, the conservation of the cathode binder harmed the flotation selectivity.

- With the EHF route, most of the active particles were individually liberated. Nevertheless, the following drying step leads to anode binder rehardening, forming unselective agglomerates. If a wet screening is performed right after the EHF process, the foils and active particles can be further separated, decreasing the content of foil impurities in the BM. Therefore, this method showed potential, provided that the recycling process is designed to remain under wet conditions.

- With the TM process, the Al foils were oxidized, corroded, and sometimes melted, which increased their fragility. These mechanisms scattered the Al foil and became a contaminant in the separation process despite binder decomposition. Therefore, it might be recommended to first allow the mechanical process to produce a BM with a low amount of Al foil, which is then pyrolyzed, reducing the amount of fine Al impurities. In addition, the pyrolysis stage decomposition of the binders leads to a difference in wettability between the active particles in the fine fraction of the BM. Hence, this TM route is the preferred route for a selective separation of the graphite and LMO particles.

By using automated mineralogy, this paper showed the unique influences of three recycling operations, revealing their strengths and weaknesses, which could be controlled, avoided, or improved to increase the LIB recycling efficiency. This work also demonstrates that only by understanding the relationships between the design of cell components and recycling it is possible to identify the inherent challenges posed by the existing battery designs, thus highlighting the design-for-recycling needs for future battery technologies.

ASSOCIATED CONTENT

Supporting Information
The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsestengg.2c00177.

Dissolution of the binder; and major phases grouped according to the LIB components (PDF)

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Funding
The authors gratefully acknowledge the Helmholtz foundation for base funding within the PoF III (project-oriented funding part III) for the BooMeRanG project. A.V acknowledges her recent funding through the BMBF Competence Cluster Recycling and Green Battery (GreenBatt) within the project “ecoLiga” (03XP0326B). R.S.G. acknowledges the BAT-Circle2.0 project (Business Finland grant number 44886/31/2020).

Notes
The authors declare no competing financial interest.

ACKNOWLEDGMENTS
The authors would also like to thank Accurec GmbH for pyrolyzing the batteries and UVR FIA GmbH for grinding the pyrolyzed black mass and the XRF analysis. The authors thank Roland Wuerkert, Michael Stoll, and Sebastian Thormeier from Helmholtz Institute Freiberg for the epoxy block preparation, Simon Obando Sierra and David Guzman Gallo for their help with sample preparation, and Alejandro Abadias Llamas for his help with the flowsheet design.

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June 23, 2022

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September 26, 2022