A qualitative assessment of Lithium Ion Battery Recycling Processes

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Abstract

With the widespread adoption of e-mobility, there are high numbers of lithium Ion batteries (LIB) entering the waste stream. It is imperative that disposal and recycling strategies are developed and implemented. There is an urgent need for safe, environmentally friendly and economically affordable disposal routes for End of Life (EoL) LIBs. This study has looked at 44 commercial recyclers and assessed their recycling and reclamation processes. A novel qualitative assessment matrix termed "Strategic materials Weighting And Value Evaluation" (SWAVE) is proposed and used to compare the strategic importance and value of various materials in EoL LIBs. The sustainability and quality of recycled material are assessed by comparing the final form or composition after the recycling processes, the industrial processes and the industry type (primary sector, manufacturer or recycler). SWAVE is applied to each company, producing a score out of 20, with a higher number indicating that more materials can be recycled. The separation processes and resources from six of the prominent recycling companies are discussed further. The majority of recyclers use one or more of mechanical treatment, pyrometallurgy, or hydrometallurgy, concentrating upon high value metal extraction rather than closed-loop recycling of the metals or component materials, highlighting an environmental and technological gap. To improve the current circular economy of batteries reuse and repurposing of materials (closed-loop recycling), instead of purely recycling or recovery of metals should be considered for further development. Further studies of environmental trade-offs from recycling or recovering one material in preference to another is required.

Keywords:

Lithium ion batteries; circular economy; recycling; waste management; industrial recycling processes; comparison

Highlights:

- Qualitative measure assessing strategic importance of materials in Li-ion batteries
- Propose Strategic Materials Weighting And Value Evaluation (SWAVE) for technologies
- Companies assessed on materials recycled, and the fate of resource streams
- Most current industrial recycling processes focus on high value metal extraction
- A circular economy gap could be addressed via closed-loop recycling of more components

1. Introduction

The transport sector has been considered to be one of fastest growing sources of environmental emissions (Sims et al., 2014), contributing to more than 28% of global greenhouse gas (GHG) emissions. Moreover, this sector is one of the main sources of cities' airborne emissions that have local impacts on human health and ecosystem quality within city borders (Rajaeifar et al., 2019). It has been estimated that the global energy demand for the transportation sector will increase drastically by 30% between 2014 and 2040. Therefore, the decarbonisation of the transport sector is necessary to achieve a global 2°C average temperature target (Santos, 2017), and electrification of the transport sector is a promising path to de-carbonization of the mobility sector and reduction of airborne emissions in densely populated areas (Hill et al., 2019). Lithium ion batteries (LIBs) are an important ingredient in EVs and are already widely used in different applications from smart phones, laptops, and other portable devices. The 30 years record of their existence has shown an increasing trend in the market volume with a decreasing trend in the price specifically from 2010 onwards (Tsiropoulos et al., 2018). The global market size of LIBs in 2018 was about 160 GWh with a value of US$ 31 bn (Pillot, 2019). Projections also show that the global LIB cell manufacturing capacity is estimated to increase by four to six times by 2021-2022 compared to 2017 levels (Tsiropoulos et al., 2018). It is estimated that EVs will increase from 15,000 new EV registration of EVs in 2017 to between 84,000-500,000 registration by 2025 in the UK alone (Skeete et al., 2020). Others predict that across the EU some 7 million EVs will be registered annually in 2025 (Baars et al., 2020; Hill and Bates, 2018). Indeed the International Energy Agency predicts that the global electric vehicle stock will grow by 36% per year and by 2030 reaching 245 million EV stocks across the world (IEA, 2020). This is similar to the scenarios given by the Taskforce 40 on Critical Raw Material for Electric Vehicles which assumes a 30% year on year growth globally as its midpoint scenario (HEV TCP, 2020). Although all the above-mentioned projections do vary and are all uncertain, the upward trajectory is not. Thus the future needs for safe, environmentally, and economically affordable disposal routes for LIBs are even more apparent, as LIB usage is increasing in all sectors. In line with that, LIB recycling is becoming more prevalent. Recycling LIBs could help in securing raw materials supply for EV batteries, reducing the high amount of energy use and environmental emissions from EV battery life cycle (Rajaeifar et al., 2020). Therefore it is imperative that the sourcing of the materials to manufacture these batteries are also tied into what happens to the materials at the battery end of life, thereby following a circular economy approach making the best use of strategic elements and critical materials (Baars et al., 2020).

1.1. LIB end of life and recycling options

There is a significant body of literature which reviews the recycling of LIBs from different perspectives.

All agree that many more development are required to ensure that economical and sustainable options are available for complicated battery system. Gaines (2014) carried out a thorough review of automotive battery recycling based on existing Lead-acid battery (PbA) recycling, to develop a sustainable LIB circular economy. They concluded that several future technical, economic, and institutional roadblocks need supporting actions to guarantee viable solutions at the batteries' end of life. In the US the recovery rates of Lead-acid batteries are 99 % (Gaines, 2014), partly because Lead-acid and Ni-Cd recycling has been encouraged due to environmental and health hazards with government legislation for recycling. Lead-acid batteries are an example of a mature technology with a well-established recycling practice. EU legislation in 2006 set minimum collection rates for Lithium-ion portable batteries in member states. These rates were 25% by September 2012 and 45% by September 2016 (European Union, 2006). Despite not being a member of the EEA (European Economic Area), Switzerland has adopted similar legislation and achieved a portable battery collection rate of 71 % in 2015 (Perchards and SagisEPR, 2016). Twelve EEA countries had already achieved the collection rate of 45 % by 2015 (Perchards and SagisEPR, 2016). Sweden leads the way within the EEA, collecting the largest total weight of portable batteries per capita (around 350 g per year since 2012), and is the only country for which a clear delineation by battery chemistry can be obtained, showing that LIBs have the lowest return rates of any battery chemistry (Perchards and SagisEPR, 2016). The UK collection rates for portable batteries in 2016, 2017 and 2018 were 44.95 %, 44.89 %, and 45.23 % respectively (Environment Agency, 2019). The disparity between recycling rates of PbA and LIBs has been attributed to the uniformity of the Lead-acid battery chemistry. The PbA chemistry and construction are remarkably simple and therefore allow for easy and cost effective recycling (Heelan et al., 2016). In comparison, although PbA currently has a larger market share the value of the components in a LIB is considerably higher, however due to complex battery structures and the lack of standardised geometries and chemistries specifically within the positive electrode (cathode) (as illustrated in Figure 1), which is often the most valuable component, recycling and reclamation are far more challenging and expensive (Gaines, 2014; Heelan et al., 2016). Wang et al., (2014) have modelled economies of scale for future LIB recycling infrastructure to analyse the profitability of battery recycling. Here it is highlighted that the mix of cathode chemistries available in the waste stream and the resultant material mass and value extracted by the process are the most important factors. For example the variability in recycling profit can be between $860 per metric ton for Lithium Manganese Oxide (LMO) cathode batteries to $8900 per metric ton for Lithium Cobalt Oxide (LCO) cathode batteries. It should also be noted that due to expected economic revenue, most current recycling processes have focused on reclaiming the cathode materials from spent LIBs and less effort has been made to recycle other materials (Harper et al., 2019; "Recycle spent batteries," 2019).

1.2 Strategic Elements and Critical Materials

Strategic elements and critical materials are of high economic importance and at risk of low supply, or supply disruption. Figure 1 shows the percentage mass of various components in a LIB. Of the main materials in a LIB, cobalt and graphite were considered both sufficiently vital and vulnerable to supply risk by the European Commission in 2017 (European Commission et al., 2017). This was joined by lithium in the 2020 report (Blengini et al., 2020). Recycling of strategic elements and critical materials is essential to reducing the environmental and humanitarian impacts of these materials, and can contribute to reducing reliance on suppliers which are prone to disruption. Therefore the primary material reclamation is extremely important for future sustainability.

Much of the focus in LIB development is in changes to the chemistry of the cathode active material. The values of the cathode metals are highly variable with time, with cobalt reaching $95,000/Tonne in March 2018 and dropped to $29,000/Tonne by March 2019 on the London Metal Exchange (Exchange). Currently Lithium- and nickel-rich $Li[Ni,Mn,Co]O_2$ (NMC), and $Li[Ni,Co,Al]O_2$ (NCA) are emerging as popular chemistries for the latest EVs, due to their higher specific- energy densities, and lower cobalt content (Chen et al., 2019). Cobalt is primarily sourced from the Democratic Republic of the Congo, which owns 46% of global reserves, and provides 69% of the EU cobalt (Mancini et al., 2020). Concerns have been raised about the use of child labour, and artisanal mines (Mancini et al., 2020; Nkulu et al., 2018). It should be pointed out that in these low income countries mining may be the main source of income for families, and consideration towards education, regular income and access to responsible markets is required (De Brier et al., 2020). As the industry moves to reduce the Cobalt content of cathode materials, and utilise more Ni, Mn or Fe, the reclaimed value of the transition metals are expected to drop further, and other short loop recycling methods need to be investigated to lower the reuse, recycling or recovery costs. Depending on the Circular Economy Strategy (from technology driven to policy driven policies that support recycling), between 30% to 90% of the total demand of cobalt for LiB can be supplied by 2050 from recycling (Baars et al., 2020). In its scenarios, the IEA hybrid and electric vehicle technology collaboration programme suggest that by 2030 250 ktons of Li, 260 ktons of Co and 1,300 ktons of Ni will be needed annually for EV batteries (HEV TCP, 2020). With the projected increase in the consumption of lithium in automotive battery applications, and although not currently on the critical materials list, sourcing will need to be considered. It is predicted that recycling of LIBs with an intent to recycle 90 % of the lithium will become necessary to meet future demands (Choubey et al., 2017; Swain, 2017).

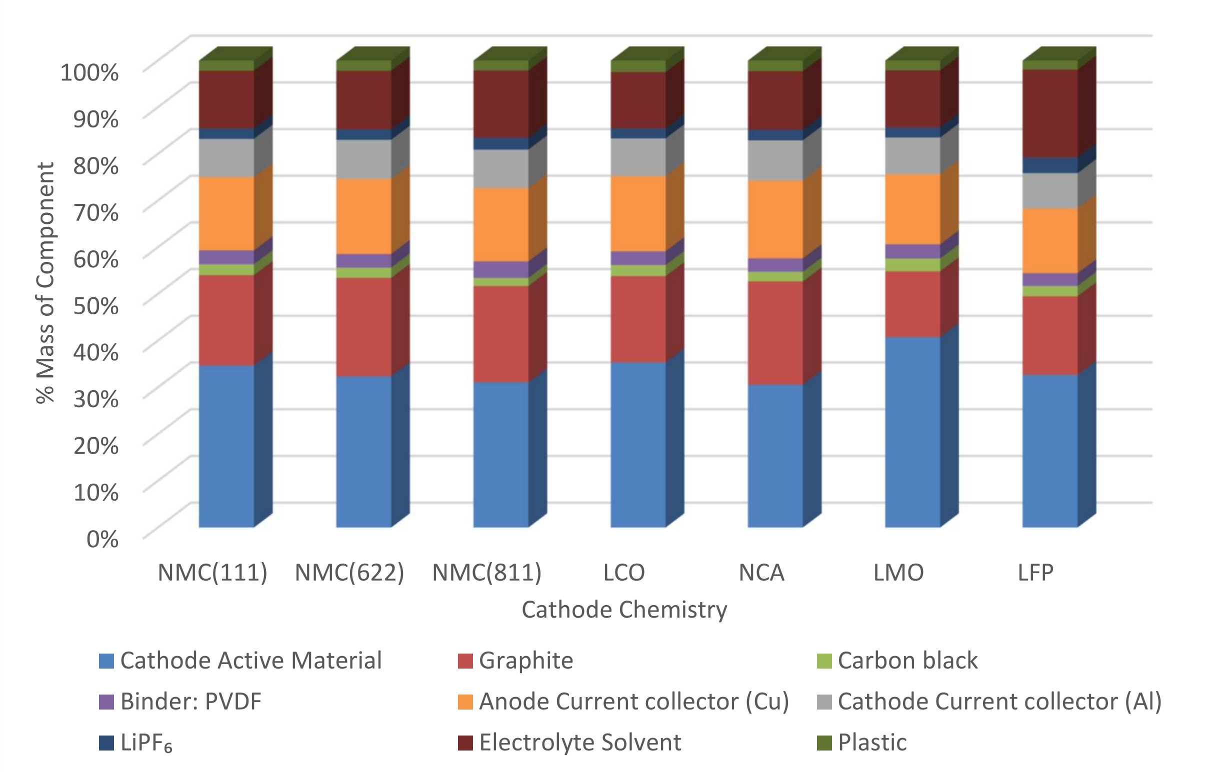

Figure 1 Relative weight percentages of different components of a large format pouch cell from an electric vehicle (Dai et al., 2019).

The majority of the world's supply of flake graphite comes from China, and 100% of the world's uncoated spherical graphite comes from China (Benchmark Mineral Intelligence, 2019). Currently, the cost of graphite is so low ($3,400/Tonne) (Benchmark Mineral Intelligence, 2019), and the required grade is so high that reclamation of graphite is not economically viable. (Moradi and Botte, 2016; Rothermel et al., 2016; Velázquez-Martínez et al., 2019). The common fate of graphite in the recycling processes is as an energy source and reducing agent in pyrometallurgical processes. However, with the classification of graphite as a critical raw material by the EU and USA, other lower cost routes such as direct recycling have been investigated (Marshall et al., 2020; Rothermel et al., 2016; Sloop et al., 2020). Studies on the economic feasibility of graphite recycling and long term reuse in a battery are now required.

1.3 Recycling Opportunities

To create a true circular economy for LIB with a hierarchy of reduce, re-use and recycling opportunities several key research challenges need addressing; automation of disassembly, safety and efficiency in dismantling, regulation of the recycling market, slag, plastics, electrolyte, and anode recycling, purity of materials waste streams, scaling up the recycling processes to industrial level and the development of new recycling processes for new chemistries and components (Chen et al., 2019; Sommerville et al., 2020; Yun et al., 2018). It can be concluded that in the future recycling of LIBs will become a necessity rather than an option mainly (but not only) due to the following driving factors: 1) high cost and unreliable supply of some critical raw materials which are needed for LIB manufacturing, 2) safety concerns regarding stockpiling or disposal of spent LIBs, 3) environmental concerns regarding disposal of spent LIBs.

Although a large number of publications reported on commercial recycling processes individually for EoL LIBs, the strategic and economic thinking of materials and the comparison of these recycling processes are still lacking in the literature. In this work, we have collated a large quantity of data from lithium ion recycling companies, of which we found information upon 60 different companies with varying quality of data. We discuss the main processes using the data from 6 companies for which a large degree of good quality data is available. These examples are used to illustrate the different commercial processes which separate the aforementioned resources and materials. We propose a qualitative assessment matrix we term: "Strategic materials Weighting And Value Evaluation" (SWAVE), which assesses the strategic importance and value of different materials in EoL LIBs. Besides, we use SWAVE to evaluate the level of 'sustainability' of commercial recycling processes.

2. Method and Materials

2.1 Recycling flow charts

We have performed an analysis of the recycling processes for 60 LIB recycling companies which take end of life batteries and for which information is available. We collected and amassed data from patents, websites, press releases, on-line resources, and from peer reviewed literature (details are given in Supplementary data). We were successful in data collection for 44 commercial operations which reported to recycle batteries and utilised this information in the SWAVE assessment. In order to understand which parts of the recycling process these companies operate in a detailed assessment of the commercial processes was first performed and the flow charts constructed. We were able to compare six of the 44 companies, for which the level of confidence in the accuracy of the data is high. These companies are Accurec, AkkuSer, Duesenfeld, Recupyl, Retriev (formerly Toxco), and Umicore. Patent databases were searched using the company names as assignees. The patents were then analysed, and the described processes were categorised. The processes have been grouped into four main functions and are discussed: stabilisation to render the cells more inert, opening and comminution to expose the interior, separation to concentrate the constituent parts of the cells into separate streams; and material extraction. The cells components were divided into eight parts: casing, aluminium foil, copper foil, cathode active material, graphite, separator, electrolyte solvent, and lithium. The path of each of these cell components through each process was then illustrated.

2.2 SWAVE Assessment Matrix

We propose a qualitative assessment of the ‘sustainability’ of the recycling processes and apply this to all 44 companies which take in end of life batteries and for which some information is available. The strategic material and critical material importance is contextualised with the sector these companies operate in, and the confidence in the information available. For ten common battery components, a Strategic material Weighting And Value Evaluation (SWAVE) has been derived based on value (USD/tonne) and criticality. Criticality data was adapted from Hayes and McCullough (2018), describing the percentage of publications since 2014 which consider each material to be critical.

3. Recycling flow flowcharts for commercial recycling processes

In the case of large assemblies of cells such as an EV pack, cells are often discharged to render them safer for handling and to recover unused energy. Packs may then be disassembled to the module or cell level for recycling. A generalised recycling loop showing the potential routes to recycle battery cathode materials is shown in Figure 2, with processes in red, and materials in blue. Large scale recycling may use a combination of pyrometallurgy, physical separation techniques and hydrometallurgy and, some recyclers only producing a “black mass” of active material which is sold on to a third party for hydrometallurgical or pyrometallurgical recovery. The processes have been compared in detail for 6 companies for which high quality information was available. Diagrams showing how the various battery components are separated are shown in Figure 3, and a simplified flowsheet which groups common separation techniques by colour is shown in Figure 4. The sustainability or resource reclamation efficiency of the recycling processes from 44 companies including the 6 detailed above has been compared in Figure 5. Comparisons were made based on the fate of the cathode material according to the flow diagram in Figure 2. Which components are reused or recycled, how valuable, strategic, or critical these materials are, confidence in the information available, which industrial sector the company operates in, and the scale of their operation is shown in Figure 5.

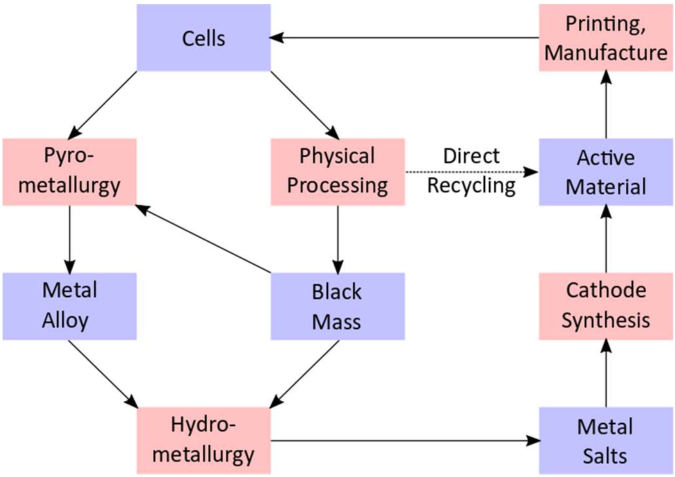

Figure 2 Generalised Recycling loop, showing processes in red, and intermediate products in blue.

3.1. Physical Processing

3.1.1. Size Separation

Size separation is the simplest form of physical separation, and is common to all the recycling techniques in Figure 3**Error! Reference source not found.** except Umicore, which treats cells whole. Duesenfeld and Recupyl use size separation to separate black mass in the form of a fine powder from coarser components of shredded battery, such as current collector foils, separator, and optionally casing materials (Hanisch et al., 2017; Tedjar and Foudraz, 2010). AkkuSer's patents describe the use of size separation in the form of filtration to remove particulates such as paper, plastics, and powders from air streams (Pudas et al., 2015). These air streams come from the comminution process. Recupyl and Retriev both apply size separation in the form of filtration, to separate solids and liquids (Smith and Swoffer, 2013; Tedjar and Foudraz, 2010). In both cases, electrolyte and a solution of lithium are separated from a mixture of anode and cathode coatings. Accurec's process is described as using a combination of size, density and magnetic separation (Meshram et al., 2014), to remove a Fe-Ni fraction, Al fraction, and Cu/Al fraction but the order in which they are used, and the materials which each process separates are not clear (Georgi-Maschler et al., 2012; Velázquez-Martínez et al., 2019).

3.1.2. Density Separation

Density separation separates materials by differences in their densities. The separator plastic utilised in LIB is commonly polyethylene and/or polypropylene, and is very low in density (<1 g cm^{-3}). The electrode foils made of Al (2.7 g cm^{-3}) or Cu (8.96 g cm^{-3}) also vary greatly in their density. A report in which Accurec was involved (Weyhe and Melber, 2016) investigated the use of density separation in battery recycling. This study demonstrated the possibility of separating casing materials from electrode foils and active material in a zig zag separator, and also demonstrated separation of Al, Cu and active material through a vibrating screen with high airflow followed by a cyclone separator (Weyhe and Melber, 2016). For AkkuSer's process, a "cyclonic air remover" removes evolved oxygen, hydrogen, and low density material such as paper and plastics, but not higher density plastics such as pouch material (Pudas et al., 2015). This low density material is added to AkkuSer's other battery recycling streams for pyrometallurgical recovery of small amounts of cobalt. In Duesenfeld's process, two density separation steps are described in (Diekmann et al., 2017), firstly to separate casing material, and after the removal of the active material, density separation is applied again, to separate Cu, Al, and separators. In both cases, zig zag separators are described. Retriev's process uses density separation in the form of a shaker table to separate the electrode foils and plastics from the comminuted cells (Dunn et al., 2012; Kelleher Environmental et al., 2019). Whilst Retriev does own more recent patents (Smith and Swoffer, 2013), which do not describe density separation, it is unclear if this patented technique has been implemented. Recupyl describes density separation to separate electrode foils from separator plastics, via a shaker table (Tedjar and Foudraz, 2010).

3.1.3. Magnetic Separation

Magnetic separation is primarily used to remove casing material. Magnetic separation is described in patents owned by AkkuSer (Pudas et al., 2015), Recupyl (Tedjar and Foudraz, 2010), and Duesenfeld (Hanisch et al., 2017). Accurec is also described as utilising magnetic separation, as part of their process to remove casings (Georgi-Maschler et al., 2012). Duesenfeld's patents (Hanisch et al., 2017, 2015) describe a magnetic separator as optional (prior to one of the zig zag (density) separators); more recent literature does not indicate it has been implemented (Diekmann et al., 2017), therefore it has been omitted in this case.

3.2. Material extraction

Following the physical separation techniques, three options are available, direct recycling, where the electrode coatings are relithiated and reincorporated into a new cell, or extractive metallurgy through hydrometallurgy, or pyrometallurgy. Only hydro and pyro metallurgy and currently performed commercially. For a detailed review of metallurgical processing routes see Brückner et al., (2020).

3.2.1. Pyrometallurgical

Pyrometallurgical recycling is one of the most ubiquitous metal recycling technologies used today. For

LIB cells or modules of sufficiently small size are put into a large furnace to produce an alloy of the most valuable metals, and is utilised by Accurec and Umicore. In the case of Umicore's facility in Hoboken, only modules or packs larger than a shoebox require disassembly prior to pyrometallurgical recycling (Treffer, 2018). In the case of LIBs, Cu, Ni, Co, and sometimes Fe are recovered as an alloy from the recycling process which is subsequently separated through hydrometallurgy (Gaines, 2018). The electrolyte, plastics, and graphite burn. Al, Li, and Mn are not generally recovered as metals, and will be found in the slag, which is commonly used as an aggregate (Gaines, 2012), though research into Li extraction from slag is ongoing (Sommerfeld et al., 2020; Weyhe and Melber, 2016).

3.2.2. Hydrometallurgical

Hydrometallurgical recycling involves dissolving the valuable cathode materials in acids, and separating the constituent metals using solvent extraction. This approach is proving more popular as more of the LIB can be recycled. It is noted that as the content of valuable metals such as Co in LIBs decrease, the profitability of pyrometallurgical recycling also decreases (Gaines, 2012). In order for hydrometallurgy to be cost-effective, it is necessary to ensure that a minimum of extraneous material is subjected to this process. Material such as electrolyte, plastics, casings, current collectors, and graphite will not be recycled by hydrometallurgical processing. In order to concentrate the cathode materials, packs or modules are safely disassembled to a manageable size, cells or modules are comminuted to produce a free flowing material, which is subjected to physical separation processes to isolate the electrode coatings as performed by Duesenfeld and Recupyl. Umicore performs hydrometallurgy after pyrometallurgy to further extract the transition metals.

3.3. Other Methods

The contemporary Retriev patents (Smith and Swoffer, 2014, 2013) describe producing a filter cake of black mass, which is heated to destroy the binder and "modify the carbon". This modification may affect the active cathode material to a greater degree than the carbon. The carbon can be removed via froth flotation, and the "heavy solids" comprising metal oxides can be filtered. The metal oxides can be made into new lithium ion cathode materials with additional Li introduced in as LiOH followed by heating. It is unclear if the froth flotation or relithiation processes have been implemented at large scale, therefore have been omitted. The Retriev process is discussed by various authors (Ekberg and Petranikova, 2015; Gaines et al., 2011; Valio, 2017; Vezzini, 2014). These authors describe how screening and shaker tables are used to separate three streams: "battery fluff" comprising plastics and casings; a finer fraction of foils; and the black mass in lithium brine. Valio (2017) discusses the importance of thermal treatment to remove or degrade the binder due to both the performance of the regenerated cathode material and to ensure an adequate difference in hydrophobicity of carbon and mixed metal oxide for froth flotation separation. The author notes that a temperature of 500 °C in the presence of oxygen is required to remove the binder without burning off the carbon from the anode. PVDF decreasing in mass at 500 °C is supported by thermogravimetric analysis (TGA) (Hanisch et al., 2015).

Other separation techniques such as eddy current separation, and electrostatic separation are discussed in literature, but the authors have yet to come across any implementation of electrostatic separation on a large scale.

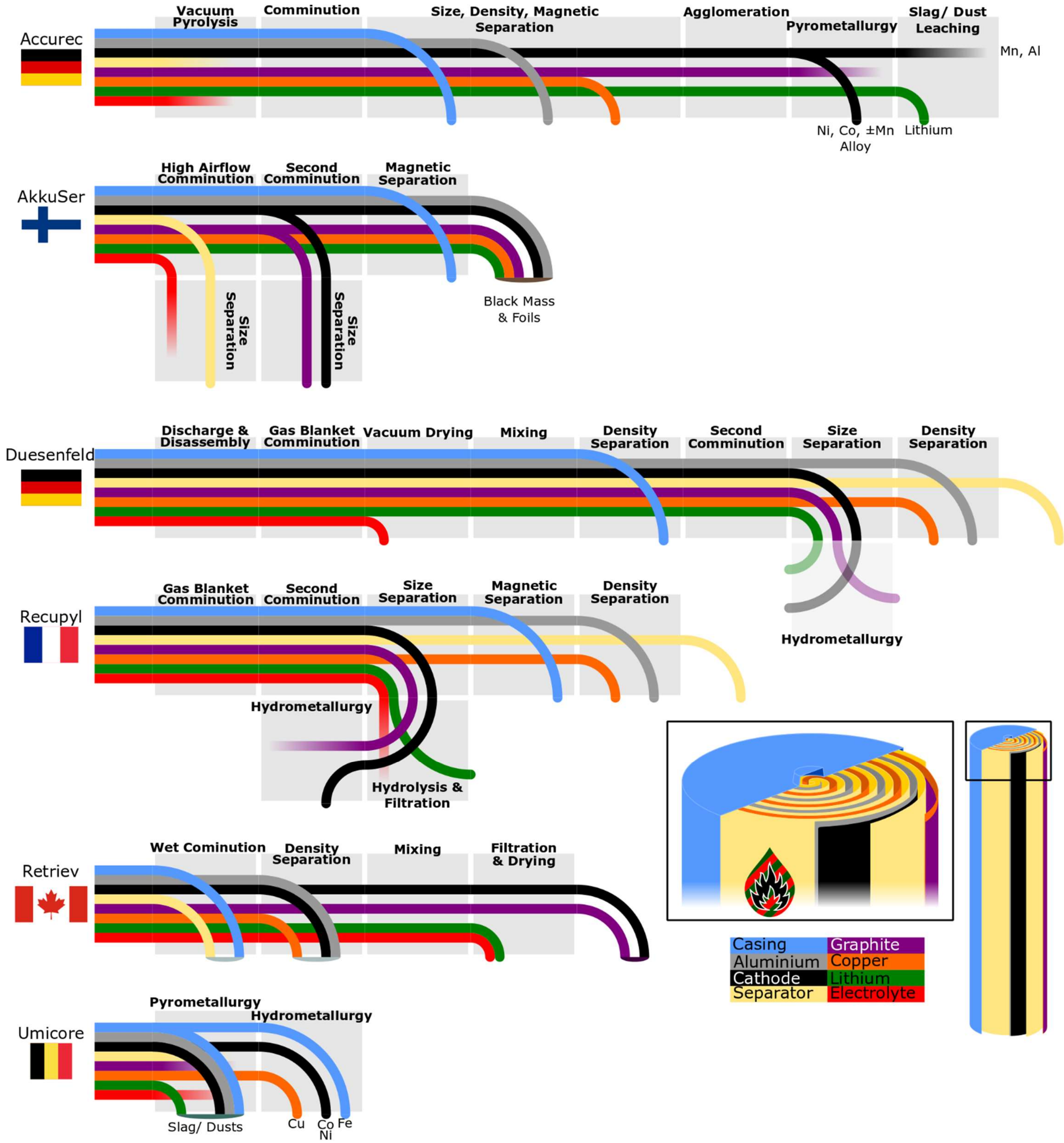

Figure 3 Summary flowsheets of processes used in six large scale recycling operations for lithium ion cells, owned by Accurec, AkkuSer, Duesenfeld, Recupyl, Retriev, and Umicore, based on patent literature (Cheret and Santén, 2008; Hanisch et al., 2017, 2015; Pudas et al., 2015; Smith and Swoffer, 2013; Tedjar and Foudraz, 2010) and (Weyhe and Melber, 2016).

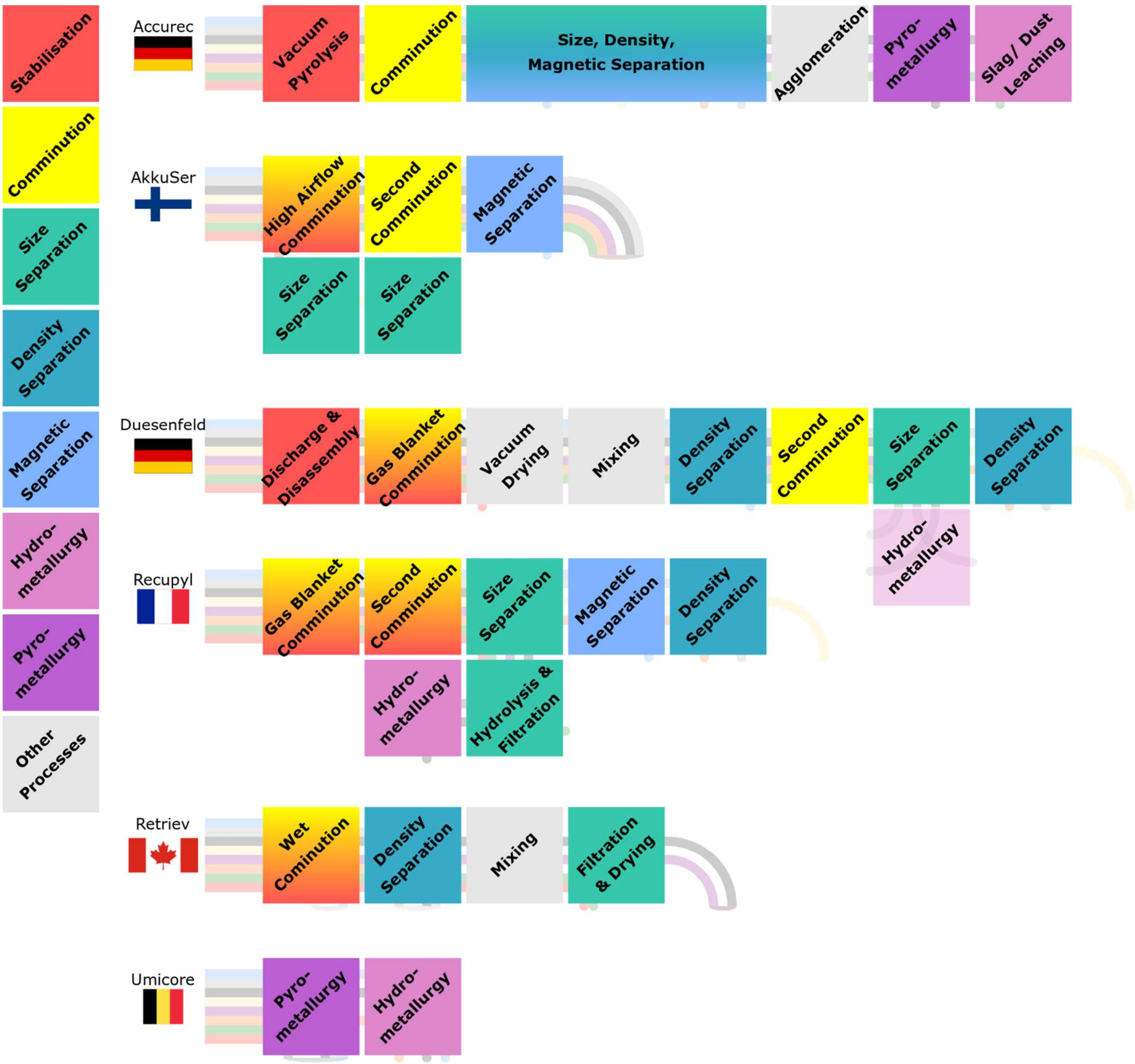

Figure 4 Processes used in recycling LIBs. Similar processes are grouped by colour to highlight similarities in these processes.

3.4. Summary of commercial recycling processes

Of the operations discussed above, Accurec and Duesenfeld use distinct stabilisation steps for processing of LIBs prior to opening the cells. Accurec utilises vacuum pyrolysis at 250 °C to remove the electrolyte and plastics (Georgi-Maschler et al., 2012), which are condensed and destined for "thermal use" (Accurec, 2018), which is not considered recycling according to EU/493/2012. Duesenfeld discharges large battery packs prior to disassembly. This is feasible for Duesenfeld, which is focussed on the recycling of battery packs from EV's, rather than individual cells (Diekmann et al., 2017). Despite this discharge and disassembly step, Duesenfeld's patents still mention the use of an environment low in oxygen and moisture (Hanisch et al., 2017, 2015) to prevent ignition of flammable components during the crushing process. This approach of stabilisation during comminution is also used by Recupyl (Tedjar and Foudraz, 2010). These "inert atmospheres" exclude oxygen and moisture, by using nitrogen or carbon dioxide (with or without argon). Carbon dioxide reacts to form a passivating layer of lithium carbonate on any metallic lithium exposed to the atmosphere during the opening process. Batrec also owns a patent (Zenger et al., 2010) which describes comminution under carbon dioxide for batteries containing lithium. Recent literature discussing Batrec's LIB recycling process cite information which can be traced back to this patent, sources which predate this patent, or Batrec's own website, which does not (at the time of writing) provide any mention of comminution under carbon dioxide. In lieu of a nitrogen or carbon dioxide blanket, AkkuSer has a unique approach of shredding under a high flowrate of air. This high flowrate of air through the shredding step prevents flammable gases from building up, and keeps the temperature at 40-50 °C (Pudas et al., 2015). Ekberg and Petranikova (2015) state that AkkuSer uses an inert atmosphere during the crushing processes, contrary to Valio (2017) who states that no inert atmosphere is used.

A Mitsubishi heavy industries patent describes crushing lithium ion batteries at -50 °C (Tanii et al., 2003). Literature commonly references "the Toxco process" when referring to prior art that advocates cryogenic pre-treatment of cells (Bernardes et al., 2004; Cardarelli and Dube, 2007; Ekberg and Petranikova, 2015; Espinosa et al., 2004; Georgi-Maschler et al., 2012; Knights and Saloojee, 2015; Lain, 2001; Meshram et al., 2014; Sonoc et al., 2015; Swain, 2017). Toxco Inc. rebranded in 2013 as Retriev Technologies Inc., retaining ownership of patents (McLaughlin and Adams, 1999; McLaughlin, 1994). The earliest of these articles by Lain (2001) stated that "the Toxco process is designed for all types of lithium containing waste". This process is well suited to lithium primary batteries, which contain metallic lithium. It has often been cited as the source of information regarding LIB recycling by Toxco (Bernardes et al., 2004; Espinosa et al., 2004). Interestingly patents (McLaughlin and Adams, 1999; McLaughlin, 1994) also describe exposing the frozen shredded or crushed material to water in order to form lithium hydroxide, and then converting it to lithium carbonate for lithium extraction. Retriev Technologies' patents US 8,616,475, US 8,882,007 (Smith and Swoffer, 2014, 2013) describe multi-stage "crushing spent lithium ion batteries under an aqueous spray". Literature on the cryogenic aspect of the processes owned by Retriev is not always explicit regarding the difference between lithium-ion and lithium primary cells, and often cites patents and literature which have since been superseded. Retriev does not use liquid nitrogen in their lithium-ion battery recycling process (Coy, 2017; Kelleher Environmental et al., 2019). Cryogenic processing is considered a greater potential safety risk than stabilisation by discharge as the electrochemical energy has not been removed from the cell, the rate of reaction has merely been slowed.

Comminution is utilised to provide a free flowing material for downstream separation processes. This is common to all hydrometallurgical recycling processes, and processes which produce black mass for processing by third parties. Patent literature commonly describes “crushing” (Hanisch et al., 2017; Pudas et al., 2015; Tedjar and Foudraz, 2010), however, some patents specify the use of shredders and hammer mills (Smith and Swoffer, 2013; Zenger et al., 2010). Comminution may be applied repeatedly in one recycling process. This is explicitly mentioned in patents owned by AkkuSer, Duesenfeld, Recupyl, and has been studied by projects affiliated with Accurec (Weyhe and Melber, 2016). Multiple stages of comminution are utilised as comminution by many orders of magnitude in one operation is less efficient than utilising two comminution steps in series, where different conditions can be applied in each step. Duesenfeld removes casing materials between the two comminution steps. If the casing materials have been adequately liberated from the foils and black mass, this is especially useful, as it reduces the power consumption of the second comminution step. After comminution has been achieved, the free-flowing material can be put through physical separation processes. An alternative to the comminution is manual disassembly. The main problem with manual disassembly methods is that they are difficult to scale up in comparison to automated battery grinding or shredding processes (Granata et al., 2012), however, such processes are under investigation (Marshall et al., 2020), and some companies appear to be disassembling cells manually (*Company Profile Video Anhua Taisen Recycling Technology Co. Ltd.*, 2018).

4. Results and Discussion

Most of the focus from the companies evaluated above is upon the valuable metals such as copper, nickel and cobalt. Companies from the primary sector commonly reclaim these as a metal alloy which can then be transferred to other sectors to further separate into the component metals in a symbiotic relationship between recycling and the primary sector, which is difficult to capture. Umicore, a recycler who makes active material, co-processes LIBs with other wastes, and has integrated these processes but only reclaims metals from pyrometallurgical and hydrometallurgical processes, therefore the SWAVE score is relatively low at 7.5.

For SWAVE the following assessment and sources were used; the component prices are sourced from London Metal Exchange (The London Metal Exchange, 2020), Shanghai Metal exchange (SMM Information & Technology Co Ltd, 2020), and Alibaba websites on 15th of September 2020, and Benchmark Mineral Intelligence (2019). Each component is given a starting weighting of 1, which is increased by 1 for a criticality value ≥ 40 %, and/or increased by 1 for a price ≥ \$3,000/tonne.

Aluminium was given an assigned weighting of 2, provided that Al was recycled as a metal, rather than recovered as a compound, due to the large energy investment in Al production.

Component	Criticality (%)	USD/Tonne	SWAVE	Price Source
Li	**40**	**7,250**[a]	3	LME
Co	**73**	**33,000**	3	LME
Ni	0	**15,090**	2	LME
Mn	**40**	1,565	2	SMM
Cu	25	**6,788**	2	LME
Al	33	1,582	2[b]	LME
Fe	25	298	1	LME
Graphite	**63**	**3,400**	3	Benchmark
Solvent	n/a	<2,000	1	Alibaba
Plastics	n/a	<1,000	1	Alibaba

Table 1 A comparison of criticality and price of various battery materials, and the SWAVE applied to each component. Criticality data is from Hayes and McCullough (2018), describing the percentage of publications after 2014 which consider each material to be critical. The Strategic Weighting and Value Evaluation was increased by 1 for each material with a criticality ≥ 40, or price ≥ $3,000/tonne. [a] Price for Li_2CO_3, [b] Al was given an increased weighting, provided it was recycled as Al metal rather than Al compounds.

As it was not always possible to acquire reliable information from companies regarding which components they reuse or recycle, each component listed in Table 1 was given a "likelihood of reclamation" for material from each company, ranging from "Not reclaimed", "Unlikely", "Probably", and "Reclaimed". The data is then plotted in Figure 5, with the data point as "Reclaimed" + "Probably", with error bars showing a minimum of "Reclaimed" and a maximum of "Reclaimed" + "Probably" + "Unlikely".

For example, Accurec, a German battery recycler, uses vacuum pyrolysis to stabilise cells, which are then shredded, and put through magnetic and size separation processes to remove steel casings, Al foil, and Cu foils. The remaining black mass is put through an electric arc furnace which uses graphite as a reducing agent, then hydrometallurgically processed to extract Ni, Co, and Mn. The flue dusts from the furnace have been studied for Li extraction. It is believed that Li extraction is probably implemented on a large scale. It is unclear if the solvent reclaimed from vacuum pyrolysis is recycled, it is presumed to be burned for energy recovery. From this information, the components considered to be "Reclaimed" (SWAVE in parenthesis) are: Co (3), Ni (2), Mn (2), Cu (2), Al (2) and Fe (1) with a total of 12. Li (3) is considered "Probably reclaimed". Electrolyte Solvent (1) is considered "Unlikely to be reclaimed". Graphite (3) and plastics (1) are considered "Not reclaimed" with a total of 4. This gives

Accurec a score of 15 (“Reclaimed” (12) + Probably reclaimed” (3)), with a minimum of 12 (“Reclaimed”), and a maximum of 16 (“Reclaimed” (12) + Probably reclaimed” (3) + “Unlikely to be reclaimed” (1)).

Industrial Sector and scale of operation is considered. The companies which recycle LIBs, LIB production scrap, or accept these materials as feedstocks for their processes were divided into four categories:

- Specialised recyclers for batteries, LIBs or e-waste
- Primary Sector (e.g: mining companies)
- Primary Sector companies with a dedicated LIB recycling process
- Cell Manufacturers

The Status of each company was also recorded in Figure 5 as Commercial (●), Pilot Scale (◆)Lab Scale (▲), or Planning (▼).

Additional companies are shown in supplementary materials Table 2**Error! Reference source not found.**. This list encompasses companies who take in LIBs but do not open or damage cells, such as battery collectors, sorters, and those who re-certify batteries for 2nd life applications. This table also includes recyclers and manufacturers about whom insufficient information is available, such that they could not be incorporated into Figure 5 and companies who no longer operate (such as AEA), but frequently feature in literature.

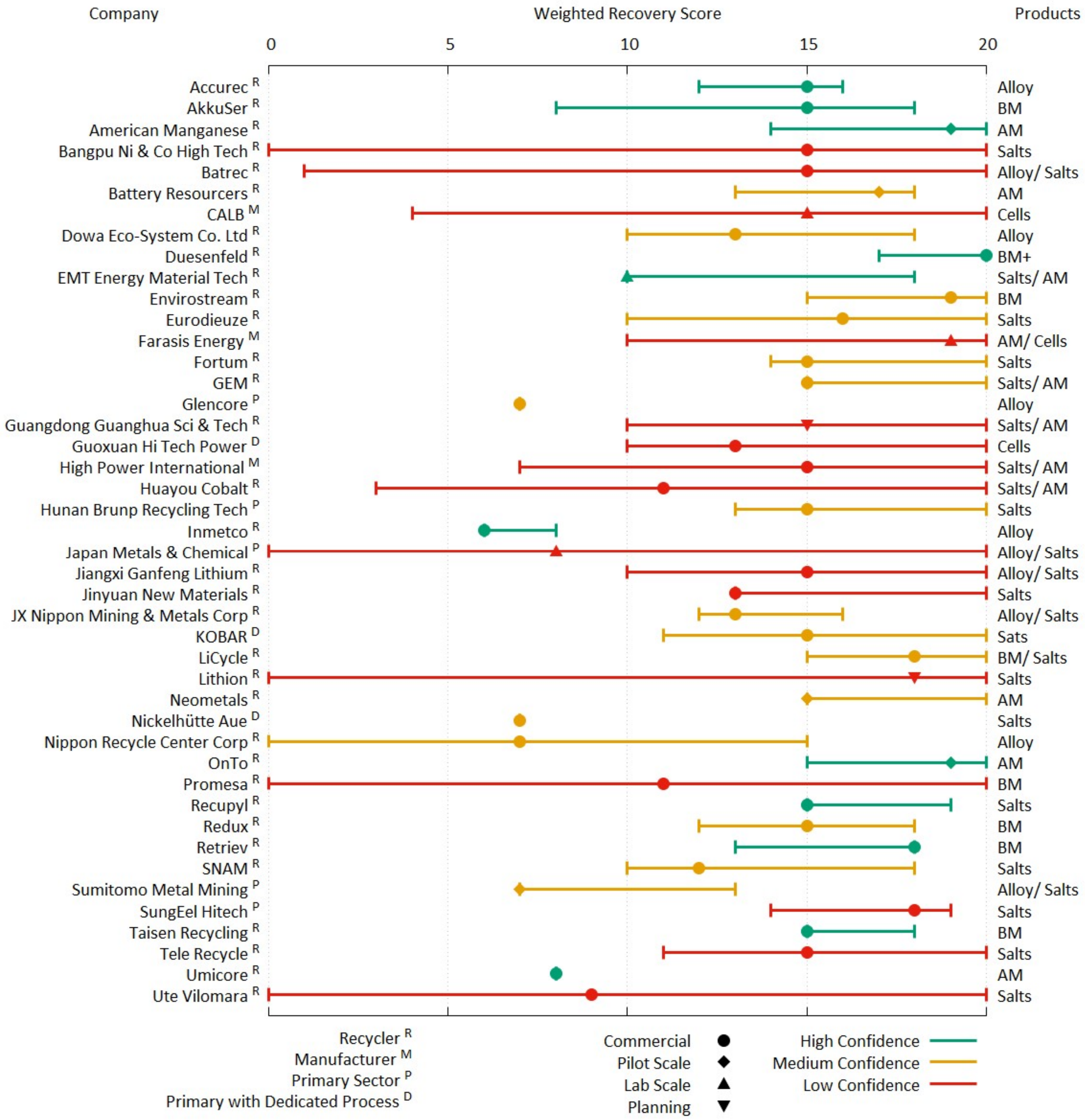

Figure 5 Total SWAVE score per company from Table 1, cathode products, sector and scale of lithium-ion battery recyclers. Black Mass (BM), Active Material (AM).

Shredding and sorting of the cell components prior to further chemical-based extraction is well documented, and is likely the major current process for effective recycling. However, there is evidence of emerging disassembly routes from companies such as Taisen (*Company Profile Video Anhua Taisen Recycling Technology Co. Ltd.*, 2018). The black mass reclaimed from this refining process is further processed using pyrometallurgical and hydrometallurgical routes. Accurec, Dowa Eco-System, and High Power International smelt the black mass, and the reclamation of manganese is also possible from this process, however, only Accurec have reported this as reclaimed metal. In terms of lithium reclamation this is primarily performed by the companies that have hydrometallurgical processes, such as Duesenfeld and Lithion, reflected in the higher SWAVE scores (18-20), or pyrometallurgical companies who may leach lithium from slag, like Accure (15). Other pyrometallurgical processing routes have lithium-manganese based waste streams which are utilised in other industries such as cement. There is surprisingly little information about the reclamation of graphite and it is assumed that for the majority of these processes it is utilised along with the plastics as a fuel or reducing agent for the pyrometallurgical processes. AkkuSer, Envirostream and Licycle reclaim black mass which is a combination of the anode (graphite) and cathode; this is shipped off for further processing by hydrometallurgical companies such as SungEel HiTech (19). However, there is the possibility of further separating the black mass to reclaim the graphite and layered oxide constituents. The reclamation of the plastics is not widespread; whilst many companies separate plastics, it is unclear if any send them for recycling rather than disposal. Duesenfeld is one of the few companies to explicitly describe the fate of plastics as disposal, or repurposing in construction. Plastics are very low value and make up a very low mass percentage of the total cell, therefore it is unlikely to make economic sense to recycle. The only recycler to report to currently reclaim solvent is Duesenfeld, where they heat the shredded cells to remove any volatile electrolyte, however the specifics of what happens to the solvent once extracted is not clear. It should be noted that this process does not, however, remove any ethylene carbonate (EC) which is contained in some cells. Accurec also separates solvents and plastics from vacuum distillation but potentially assign these for thermal use, which is not determined as recycling by the EU. Duesenfeld is currently reclaiming more components than any other recycler, and therefore have the highest SWAVE score of 20.

There are some limitations to this model and we would like to note that the companies whose final product is black mass are treated favourably, as all electrode coatings are considered to be "recycled", because the buyer of the black mass is unknown. If the buyer is a smelter, they may not recycle graphite and manganese, whereas a hydrometallurgical buyer may. Binders and conductive additives are ignored by this model, as they represent a small component of a cell (<10% by weight of the black mass) (Marshall et al., 2020), and no commercialised processes for their recycling are known. Scant information is available on reclamation of P from LFP or $LiPF_6$ on industrial scales, and this has not been tracked. It is assumed that all anodes are graphite, and all cathode chemistries are NMC, LMO, LCO, NCA, or LFP, or are otherwise primarily Li, Ni, Co, Mn, or Fe. In the case of NCA, the Al component is small (e.g: 5%) (Marshall et al., 2020), and ignored. Further details and calculations of the SWAVE scores and source information can be seen in supplementary data.

5. Conclusions and policy recommendations

Due to the widespread adoption of lithium-ion batteries in e-mobility and consumer electronics, high numbers of batteries are entering the waste stream. These streams contain strategically important elements and critical materials. There is an urgent requirement to make sure that these materials can be reused or recycled, to aid the supply of these materials to the future battery manufacturing industries, reducing both cost and waste.

In this work, we have compared information from 44 commercial recyclers and assessed their recycling and reclamation processes. A novel qualitative measure termed “Strategic materials Weighting And Value Evaluation" (SWAVE) is proposed and used to compare strategic importance and value of various materials in EoL LIBs. It should be noted that the complete data for this analysis is difficult to obtain, and the processes and routes are still being developed by many companies. The necessity of commercial protection and IP has therefore made analysis and development of this work difficult in many cases. It is therefore likely that much more collaboration is needed between both academia and the battery recycling industry itself to enable stronger circular economy models to develop in an environmentally timely manner. Most dedicated recycling companies use shredding and separation prior to chemical extraction processes such as hydrometallurgy. There is often a synergy between the recyclers who reclaim the black mass components, and the primary sector companies who then convert the black mass back to the constituent metal salts. There are additional opportunities to further purify the black mass and extract the graphite and the cathode constituents in these cases. There are also cases of companies who are integrating vertically, starting as battery manufacturers, or mining companies who are co-processing end of life LIBs for their metal values. Surprisingly little is known about what happens to the graphite from hydrometallurgical processes; the major opportunity here is for short loop recycling of the graphite back into the manufacturing stream.

The qualitative assessment of the value and importance of the materials separated by a variety of recycling processes, (SWAVE), provides a useful and timely contextualised overview of the strategic materials reclamation, weighted by the industry in which each company operates and the fate of the reclaimed materials. Reusing or recycling a high proportion of the critical materials, and valuable products in a LIB is vital to guarantee a sustainable, low-carbon future and where a 100% recyclable lithium-ion battery becomes a reality. There are many opportunities to improve the current recycling processes of lithium ion batteries, and to better reuse and recycle strategic materials and critical elements:

1. Most recycling processes are developed to reclaim valuable metals and sometimes graphite but less focus on other materials, such as solvents, plastics, lithium salts and phosphorus. The solvent in particular is a large proportion of the components of the cell and needs to be

considered if we are moving towards 100% recycling of batteries. Besides, the quantity of high value metals is expected to reduce in future batteries, which also requires us to pay attention to the recycling of more components other than metals.

2. Most cells are thought to be currently hand disassembled from limited data of commercial processes. Hand disassembly may bring many health and safety issues due to the chemical constituents, which brings the opportunity for automation of disassembly. Moreover, automated disassembly is also expected to have high efficiency and can meet the requirement for industrial scale. However, the design of LIBs varies significantly with different models and manufactures, which increases the difficulty of automated disassembly. Therefore, the standardization of cell geometry and architecture is very necessary.
3. According to the waste hierarchy, short loop or direct recycling, i.e. where the active material is reused without returning to the constituent metals or salts; is always preferable compared with direct loop recycling, i.e. where the components are remanufactured from the recycled metals or salts. However, short loop recycling requires a great purity of the material waste streams, which in turn requires the adoption of more careful independent processes to remove the cell packaging and the cell components.

To conclude here are some general policy recommendation and suggestions for further research. The findings from our study reveal a surprising lack of both strategic or circular economy thinking currently operating in the market place in EOL batteries. There is no current standardisation in end of use disposal, it is suggested that this may need to change on a national basis moving forward. Companies are overlooking internationally strategic important sources of critical materials in favour of traditional reclamation of material sources and quotas. There needs to be a policy change to drive the CE forward, one where more holistic reuse and recycling options are encouraged. These should incorporate more energy efficient reclamation processes, closed loop processes with greener chemicals use. The current and future economic analysis of these recycled materials from the developing processes are required, and we expect that in some cases incentives to encourage new markets that can utilise the extracted materials are also needed. This will be aided by further studies understanding of the lifecycle trade-offs in terms of the created emissions in utilising the identified processes of the current systems. Best practice in EoL disposal can therefore be encouraged.

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Conflicts of interest

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Supplementary information

Source information for Figure 5 and Table 2 (Sommerville, Rob et al., 2020) DOI: 10.5281/zenodo.4032883

Company	Country	Industry	Status
AEA	UK	n/a	Dead
Beijing Easpring Material Technology	China	Manufacturer	Planning
Beijing Saidemi Resource Reuse	China	Recycler	Unclear
Blue Whale Materials	USA/ Korea	Recycler	Planning
Camel	China	Manufacturer	Planning
Chery Automobile Co	China	Manufacturer	Operational
Earthtech	South Korea	Recycler	Planning
ERLOS	Germany/ EU	Collection	Operational
G&P	UK	Collection	Operational
Great Power	China	Manufacturer	Operational
Narada Power	China	Recycler	Planning
Nissan	Japan	Recertification	Operational
RMC (Raw Materials Company Inc)	Canada	Collection	Operational
Sitrasa	Mexico	Recycler	Operational
Spiers New Technology	USA	Recertification	Operational

Table 2 Other companies which currently or formerly take in LIBs

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