Recycling of Lithium-ion Batteries

By

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A thesis submitted to
The University of Birmingham
for the degree of
DOCTOR OF PHILOSOPHY

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March 2024

ABSTRACT

The demand for electric vehicles has greatly increased, driven by the public's awareness of global warming and electric vehicles being part of the solution to reduce greenhouse gas emissions that are the cause of it. Although electric vehicles have been shown to be better for the environment it is important that they are treated correctly at the end of life as they contain many valuable and hard-to-extract materials within them, particularly within the battery pack and the cells therein. Recycling of cells has the potential to further boost the environmental credentials of electric vehicles (and other battery-powered devices). Recycling of cells already takes place and more processes are being developed, however, more work is still needed. The work presented in this thesis focuses on understanding how we can move towards zero-waste recycling processes. To understand how we recycle lithium-ion batteries, we need to understand the current composition and construction of the different cells, and if we are to improve our materials reclamation yield, and if there is any scope for an improved design for disassembly.

Chapter 4 characterises three cell types: cylindrical (end-of-life samples), pouch (production scrap samples) and prismatic (end-of-life samples). Indicative of the cell types in the current market. A complete cell tear-down and cathode materials evaluation is performed with a critique and discussion of how improvements can be made for design for disassembly and recycling. This is the first time that a range of commercial cells have been compared using the same suit of techniques, and their recyclability has been the focus.

In Chapter 5, this work will look at organic acids. Can these be used for the direct recycling of cathode black mass? For this to be the case the recovered black mass must have greater capacity than the End-of-life cell (107.62 mAh/g as found by preliminary testing). Otherwise, can organic acids be used as a pre-treatment step for valorisation and hydrometallurgical treatments? For this, the leaching of metals must be controlled. Depending on the desired outcome various acids were successful for example, if retaining nickel in the black mass is the goal, succinic acid is the most effective for separation. Conversely, if cobalt retention is preferred, citric acid should be used. If optimizing first-cycle capacity is the objective, acetic acid yields the best performance.

Chapter 6, the use of DMSO, (which is considered a green solvent, but still carries some challenges such as its ability to permeate the skin barrier and carry through transition metals) is investigated to further improve valorisation of the cathodic black mass using solvent delamination to enable better valorisation by avoiding leaching if possible. Stirring and ultrasonication were used to agitate the solution at a range of temperatures to find the optimum processing conditions for the release of black mass from the current collector. This is the first time DMSO has been used in the recovery and recycling of black mass, and the first time that optimisation has been carried out on this system.

ACKNOWLEDGMENTS

To say the last five years of research have been a challenge would be somewhat of an understatement. There have been many times when I wanted to throw in the towel, call it a day, and scurry away with my tail between my legs defeated, but I haven't thanks to a whole bunch of people who I will try and thank here, but I hope to not offend if I miss anyone out.

Starting with Professor Emma Kendrick, without who's, guidance, insight, support, and expertise none of this would have been possible. Thank you for taking a chance on me and taking me on. Thank you for your patience and for making time even though at times there were months when nothing worked. I know that sometimes it seemed like your guidance and advice fell on deaf ears, I promise you it did not.

Dr Rob Sommerville, whose practical guidance has stopped me from making many a dumb mistake and saved more result sets than I would like to admit. Without your guidance and practical help, the results would have looked far worse, and the experimental design would have been weaker without our rigorous discussions in the lab.

Dr Dan Reed, without whose exceptional characterisation knowledge it would have been so much more difficult to understand what was going on you have saved me more than a few hours. Ben Pye, the group's technician without you the labs would grind to a halt, I hope I solved at least one more problem than I created. The whole of the EMG group thank you for being such a supportive, fun, and welcoming group of people.

It would be amiss to not thank my family and friends without whose ever-present support this would have been 100 times more difficult. Emily Walsh for always sticking by me through thick and thin, listening to me moan when I had a bad day and broke the 5th bit of glassware in one day, I couldn't have done it without your support. Also to my parents, thank you for your ever-present support and encouragement, it meant a lot to me.

PUBLICATIONS

Publication as a result of this work:

- A. Zorin *et al*, Acid-Assisted Separation of Cathodic Material from Spent Electric Vehicle Batteries for Recycling, Metals **2023**, *13(7)*, 1276. Contributions: E. Kendrick - Supervision, Reviewing, and editing. D. Gastol - Electrochemical methods standardisation and cell construction, T. Song - XRD acquisition and analysis. A. Zorin - lead author ICP, electrochemistry, SEM, acid delamination data acquisition, and data analysis and visualisation

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- P. Zhu *et al.*, Direct reuse of aluminium and copper current collectors from spent lithium-ion batteries, Green Chem., **2023**, *25*, 3503-3514. Contribution: ICP data collection and analysis

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Acronyms

BSE back-scattered electron. 46

DEC diethyl carbonate. 8

DMC dimethyl carbonate. 8

DMSO dimethylsulfoxide. 34, 35

EC ethylene carbonate. 8

EMC ethylmetyl carbonate. 8

EoL end-of-life. 22, 34

EV electric vehicle. 22

EVs electric vehicles. 1

FEC fluroethylene carbonate. 8

ICE internal combustion engines. 1

ICP-OES Inductively coupled plasma optical emission spectroscopy. 48–51

IEA International Energy Agency. 1, 2

LiB lithium-ion battery. 3, 21

LiBs lithium-ion batteries. 1, 3–5, 19, 21

LMO lithium manganese oxide. 29

NMP N-methyl pyrrolidone. 34, 35, 67

PC propylene carbonate. 8

PE polyethylene. 8

PP polypropylene. 8

PTFE polytetrafluoroethylene. 48

PVDF polyvinylidene fluoride. 8, 67

PXRD powder X-ray diffraction. 44

SE secondary electron. 46

SEM scanning electron microscope. 46, 47

UKAEA UK Atomic Energy Authority. 4

VC vinylene carbonate. 8

VOCs volatile organic compounds. 24

Chapter One

Introduction

1.1 The Need for Electrification

The demand for lithium-ion batteries (LiBs) in electric vehicles (EVs) has surged significantly, driven by their potential to reduce greenhouse gas emissions in the transportation sector by up to 80% by 2050. This aligns with the UK Government's long-term goal to meet the targets of the Paris Agreement [1]. Despite advancements in fuel efficiency, there has been no notable reduction in transportation-related greenhouse gas emissions since 1990, primarily due to the increase in annual car mileage. With transportation contributing approximately 27% of greenhouse gas emissions in the UK and 21% in the EU, electrifying the transport sector is deemed essential [2, 3]. Electrification not only aids in lowering greenhouse gas emissions but also reduces dependence on fossil fuels for internal combustion engines (ICE). However, it is important to note that generating additional electricity to charge EVs will be necessary. According to Nordelöf et al., EVs improve greenhouse emissions only if the energy used to recharge them is produced by renewable sources [4].

As of 2017, there were 3 million electric vehicles globally. The International Energy Agency (IEA) predicts that by 2030, the number of electric vehicles on the road will reach 125 million [5]. The adoption rate of electric vehicles is not expected to decrease. Bloomberg New Energy Finance expects that by 2040, the global electric vehicle count will reach 530 million [6], which is more than double the 2030 number predicted by the IEA. These predictions are strongly influenced by governmental initiatives on a global scale. The UK Government has set a goal in 2020 to eliminate the sale of petrol and diesel cars by 2030, which has been pushed back to 2035, aiming for all new car purchases to default to ultra-low emission vehicles [7]. Other countries, such as Norway and France, have established similar targets to halt the sale of petrol and diesel cars by 2025 and 2040, respectively.

The transition to electric vehicles will be challenging and apply substantial pressure on automotive companies, compelling them to set more significant production goals to meet consumer demand. One example is Volkswagen AG, which has announced to allocate €70 billion for the development of electric variants across its entire model range by 2030 [6]. As one of the largest automotive manufacturers in Europe, the UK automotive industry holds significant prominence, with a production output of almost 1 million cars in 2023, of which 79% were exported [8]. Therefore, it is crucial for the UK to initiate innovation in the battery sector and to be at the forefront of sustainability and end-of-life management.

Like all vehicles, electric vehicles have a life span. However, it is difficult to find official statistics from governments on how many electric vehicles come to an end of life each year and, hence, how many lithium-ion batteries require some form of final process. It is widely published that once a battery is at 70-80% of its initial capacity, it is deemed to be end-of-life as an automotive traction battery [9]. This is because at this level of capacity, the battery has undergone a level of degradation (although not discussed in depth, the common degradation mechanisms are illustrated in Figure 1.1) that has pushed it to its knee point where nonlinear aging is likely to occur [10] and modeling of this aging, the battery's capacity, and hence its range becomes challenging and unpredictable and also the battery can no longer provide a satisfactory level of power for acceleration and driving. This level of degradation takes 8-10 years of service to achieve [11] unless a vehicle is involved in a crash. Assuming that after ten years, most electric vehicles are scrapped with their batteries and using the International Energy Agency report 2022 [5] the number of electric vehicles in circulation, we can assume about 100,000 in Europe, and these will be coming to an end of life, in 2023. According to the same report, the number of electric vehicles doubles annually. These vehicles, including their batteries, will have to be recycled and reused to some extent due to incoming European directives [12], namely, the EU battery passport which is discussed in section 1.2 and Figure 1.5.

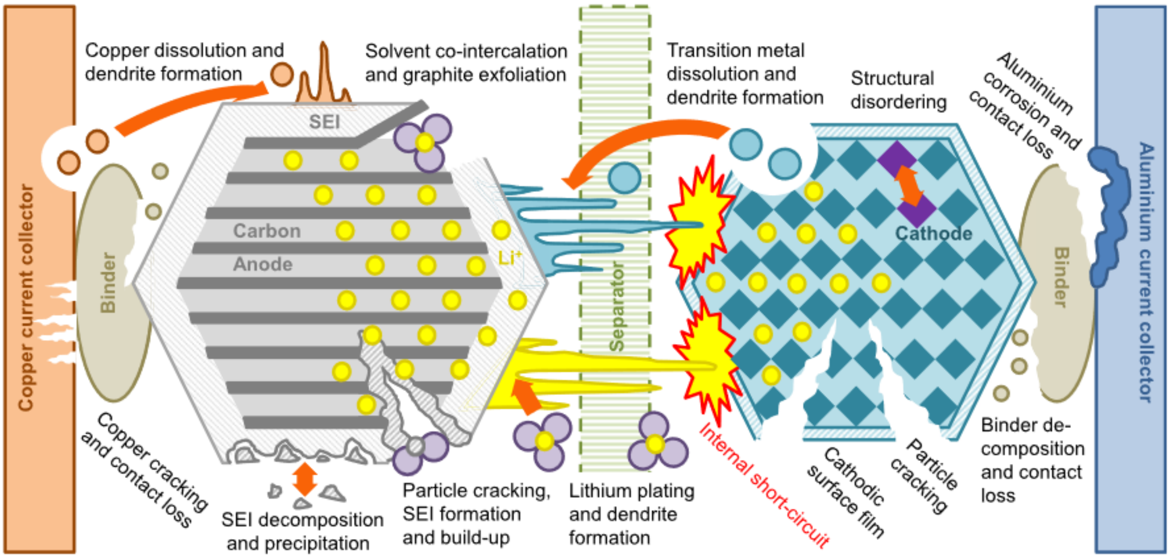

Figure 1.1: Degradation Mechanism in Lithium-ion Batteries [13].

1.2 Lithium-ion batteries

Before delving into the recycling of lithium-ion batteries (LiBs), it is necessary to understand their composition, operational principles, structure, manufacturing processes, and the materials employed. The lithium-ion battery (LiB) was first commercially introduced by

Sony in 1991, primarily targeting mobile consumer applications [14]. Goodenough, Whittingham, and Yoshino were awarded the 2019 Chemistry Noble prize for *"for the development of lithium-ion batteries"* [15]. These scientists explored intercalation materials, such as $LiCoO_2$ [16], TiS_2 [17]. The materials developed by Goodenough and conformed to the formula $A_xM_xO_2$ where A can be an alkali earth metal such as Li, Na, or K and M is a transition metal such as Co, or Ni and were patented by the UK Atomic Energy Authority (UKAEA) [18] and subsequently commercialised by Sony, continues to be utilised today. The enduring appeal of LCO is attributed to its distinctive characteristics, including a high theoretical specific capacity (270 mAh/g), and an elevated operating voltage.

However, despite the above-mentioned upsides, the application of $LiCoO_2$ is limited to smaller portable electronic devices rather than electric vehicles. This is because of factors such as cost, safety concerns related to rapid discharge, and geopolitical and environmental issues associated with the mining and use of cobalt. In terms of manufacturing geopolitical and ecological issues cause the most concern, from the standpoint of end-user safety the rapid self-discharge and thermal stability in the even of a crash cause the most concern.

Because of these drawbacks, several other cathode chemistries have become prevalent in automotive traction batteries. For instance, $LiMnO_2$ which is used by Nissan in the Leaf vehicle as a mixed cathode-material alongside $LiNiCoAlO_2$ (this will be discussed in Chapter 4). $LiFePO_4$ has been reported as being used by Tesla in China. $LiNi_xMn_yCo_zO_2$ (x+y+z=1), contains cobalt but in a lower proportion than in $LiCoO_2$. The shift to lower Co and Mn is driven by the criticality (in Europe) of Mn and Co. Although a discussion of production volumes would be interesting these statistics are difficult to find and fall outside the scope of this thesis.

Table 1.1 presents a list of the components found in lithium-ion batteries (LiBs), along with estimated costs and masses (expressed as a percentage of the entire cell) for each component [2, 19–22]. These values vary depending on cathode chemistry, binder type, solvent, and additives. The production scale and the purity of these components influence the cost. Although lithium-ion batteries can utilise other material types, such as alloy and conversion materials, this work only deals with intercalation material.

In lithium-ion batteries (LiBs), intercalation materials are initially made in a discharged state, containing lithium-ions within both the cathode and electrolyte [23]. Applying a voltage between the electrodes results in current when the battery undergoes the formation and charging process. To maintain a charge balance throughout the entire battery, lithium-ions (Li) de-intercalate from the cathode, diffuse through the electrolyte, and re-intercalate between the layers of the graphite anode, as depicted in Figure 1.2. During the discharging phase, the reverse sequence occurs, with electrons circulating through an external circuit to perform useful work. Each electrode has foil current collectors, Al for the cathode and Cu for a graphite anode [24, 25]. This is how, during the discharge portion of the operation, the battery transforms storable chemical energy into electrical energy [23].

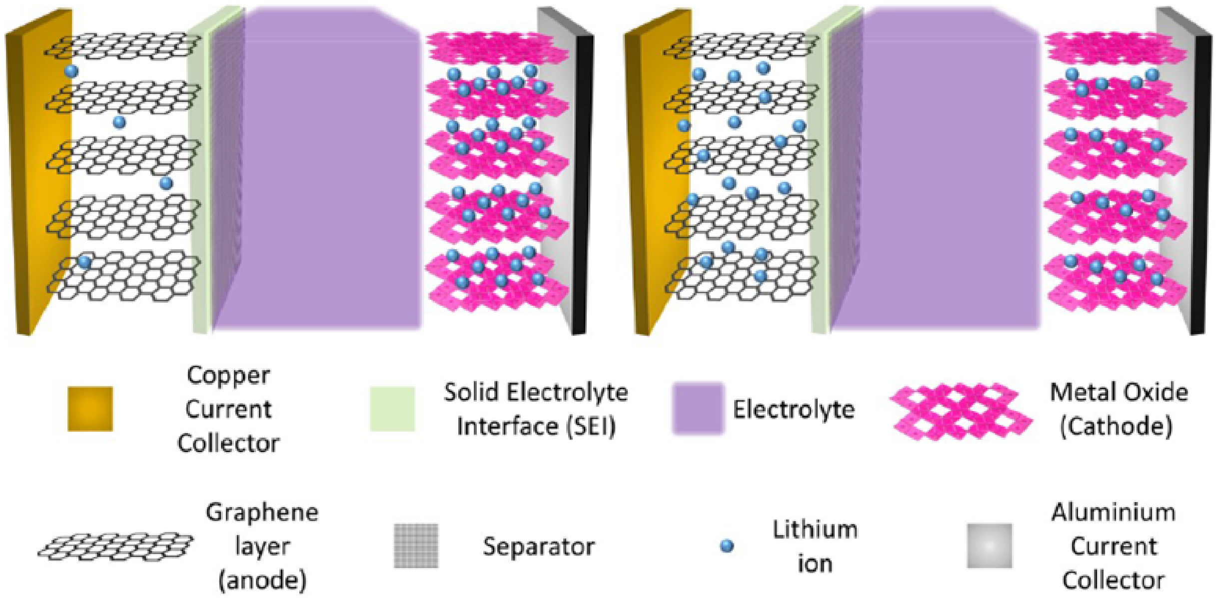

Figure 1.2: Operating principle of a LiB. Left: discharged, Right: Charged [26].

The equations below describe the redox half-reactions for the charge and discharge of a cell with respect to Li in the anode and cathode:

$$Li_xC_n(s) \rightleftharpoons xLi^+(soln) + xe^- + nC(s) \tag{1.1}$$

$$Li_{1-x}MO_2(s) + xLi^+(soln) + xe^- \rightleftharpoons LiMO_2(s) \tag{1.2}$$

Equation 1.1 describes the anodic half-reaction. Equation 1.2 shows the cathodic half-reaction (M is a transition metal). Discharge occurs from left to right, whilst charging occurs from right to left.

Table 1.1: Cell materials found in a typical LiB [2, 19–22].

Compnent	**Material**	**Composition Wt.%**	**Cost %**
Cathode active material	Layered material ($LiCoO_2$), Spinel Material ($LiNi_xMn_yCo_zO_2$), Olivine material ($LiFePO_4$)	22-25	65-70
Cathode foil	Al	4-5	1
Anode Active material	Graphite, Graphite / SiOx mix	24-26	8-9
Anode foil	Cu	3	2
Binder	PVDF	3-2	8-9
Electrolyte	EC / PC / DMC / EMC / DEC, + additives e.g. FEC / VC mix with $LiPF_6$ as conductive salt	10-12	1
Conductive additive	Carbon Black	1	0.1
separator	PE / PP	4-5	4
Cell Case	Range from metal casing to plastic to metal/plastic laminate	4-6	4

1.3 Lithium-ion Battery Design, Manufacture, and Recycling

Cells commonly come in 3 geometries: prismatic, cylindrical, and pouch. These are then connected into a module or directly into a pack in series or parallel and, most commonly, in a mix of the two to allow for appropriate power delivery and provide for an energy-dense pack. In this section, the thesis will discuss the manufacture of pouch cells; however, in Chapter 4, the thesis will discuss the disassembly and structure of all three geometries. Figure 1.3 illustrates how manufactured electrodes go from black mass components to electrodes that are ready to enter a cell. The manufacturing process is broken down into 5 stages, 1) is the slurry making, 2) and 3) are the coating and drying of both sides of the current collector, 4) is the calendaring of the coated electrode, and 5) is the cutting and tab welding. Challenges to recycling can be introduced in a number of these stages. In stage 1 the choice of binder will determine the ease with which the black mass can be removed from the current collector at the end of the life of the cell. at stage 5 the stacking and the tab geometry and fixing type (this will be discussed in Chapter 4 can determine how difficult it is to separate the various components from one another.

Figure 1.4 illustrates the cross-section of a pouch cell, where the stacked nature of a cell like this can be seen. In Chapter 4 there will be presented more schematics of how cells are constructed and stacked.

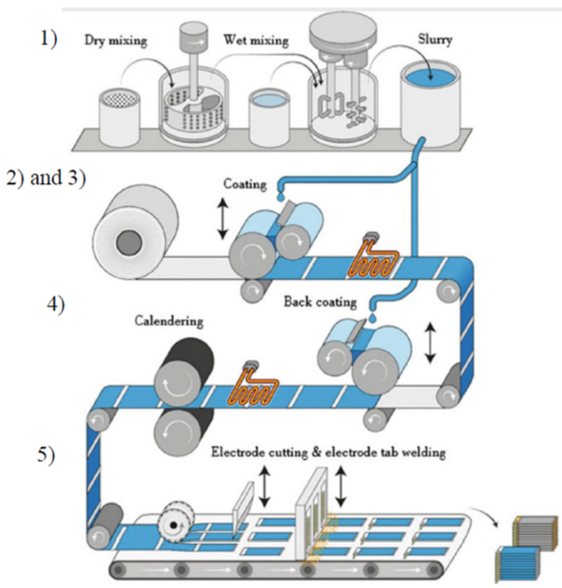

Figure 1.3: Schematic of the process of electrode manufacture [26].

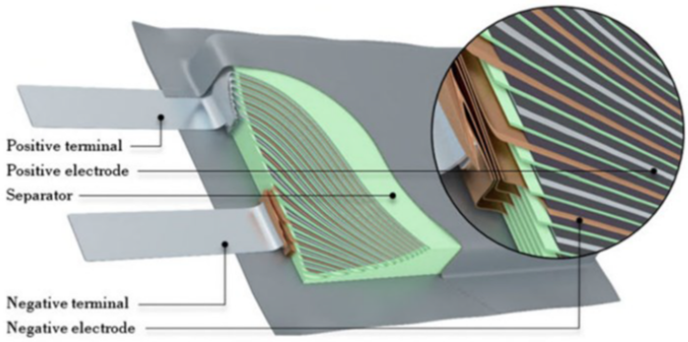

Figure 1.4: Cross section of cell structure once assembled [26].

Many governments mandate the recycling of electric vehicles. At the forefront of this is the European battery directive [12], also known as the 'battery passport' in Europe. It says all batteries must contain a unique identifier and a data sheet with manufacturing data. The directive states that by 2021, the passport must include the recovery rates of critical materials from waste batteries (Li 6%, Ni 6%, Co 16%). By 2025, the plant's carbon footprint must also be included in the battery passport and verified by a 3rd party. By 2028, the passport will have to include technical reports detailing the percentages of critical materials from recovered sources [27]. By 2030, a minimum of 70% of the cell must be recyclable. A timeline of this is also shown in Figure 1.5.

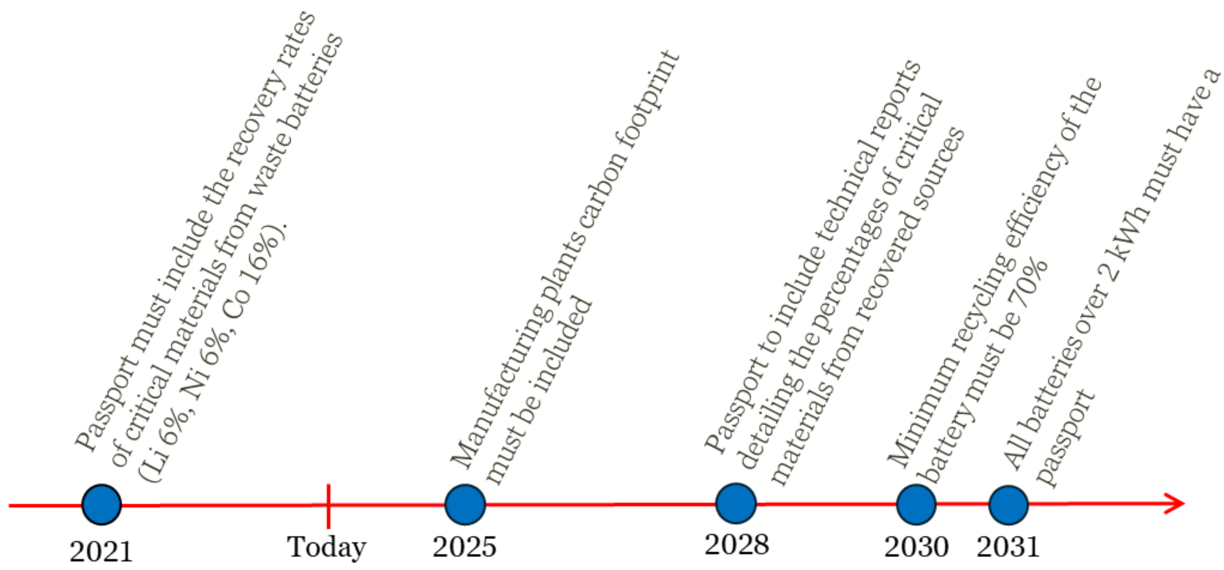

Figure 1.5: Timeline of EU directives for the battery passport [27].

Several recycling processes are currently employed worldwide, each of which has its own benefits and drawbacks. The most common are based on pyro-metallurgy with hydro-metallurgy routes, and direct recycling is a third approach that needs more research and development before it can reach industrial-scale application. Pyro-metallurgy consists of treating cells as enriched ore and placing them into furnaces with mined ore, smelting them, and extracting the most valuable constituents, such as Co and Ni. Of course, this is the most straightforward process; however, it yields the least value from the cell and loses much of the other critical materials. Hydro-metallurgy usually employs acids or alkalis to dissolve the cell components fully. The valuable metals are dissolved into solution and can then be precipitated as salts to manufacture active materials. The main drawbacks are that this can be water- and waste-intensive, and the dissolution of all metals means that some less valuable but still critical materials are not recovered, which is financially prohibitive. Direct recycling is any process where the feedstock does not undergo a phase change (i.e. battery active material always remains active material). This has the potential for the highest value recovery of material. However, it also requires the most complex processes and still needs some R&D to reach wide industrial adoption. The literature review will discuss these more thoroughly (Chapter 2).

This thesis explores methods that allow for progress towards zero-waste processes. To be able to better recycle, we need to know the starting composition and structure of the various feed-stocks available, as well as to identify potential improvements in materials reclamation and design for disassembly. Chapter 4 examines three types of cells: cylindrical, pouch, and prismatic, which are the three most commonly available cell geometries in today's market. In this chapter, a method for a full and fully traceable quantitative disassembly of these cells is presented, along with a critique and discussion on ways to improve the design for disassembly and recycling.

In Chapter 5, the optimisation of cathode material for direct recycling is performed using an acid delamination process. The aim was to retain as much value in the reclaimed cathode as possible, improving the valorisation. Through the choice of organic acid and process optimisation, lithium, and transition metals were retained in the recovered solid and used for direct or short-loop recycling, limiting the metal leaching into the aqueous waste stream.

Chapter 6, separation from the current collector of the cathodic black mass is investigated using solvent delamination to enable direct recycling or maximum value retention by not leaching transition metals from the cathode material.

In Chapter 7, the key aspects of black mass separation conducted in Chapters 4- 6 are summarised, and thoughts about future work are discussed.

Publications as a result of this work include:0

- A. Zorin *et al.*, Acid-Assisted Separation of Cathodic Material from Spent Electric Vehicle Batteries for Recycling, Metals **2023**, *13(7)*, 1276. Contributions: E. Kendrick

- Supervision, Reviewing, and editing. D. Gastol - Electrochemical methods standardisation and cell construction, T. Song - XRD acquisition and analysis. A. Zorin - lead author ICP, electrochemistry, SEM, acid delamination data acquisition, and data analysis and visualisation

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Chapter Two

Literature Review

2.1 Introduction to Recycling Processes

Several recycling methods have already been established and commercialised. These all fall along the waste management hierarchy shown in Figure 2.1. The hierarchy in order of preference is direct recycling, hydro-metallurgy, and pyro-metallurgy because each subsequent route retains less of the value of the original active material and requires more post-processing before they can be reused in a battery. The waste hierarchy differs slightly from general good environmental practice as it does not include reduction and reuse of material, because it only deals with material once it is classified as waste. Although outside the scope of in-depth discussion in this work, a reduction in the use of batteries and the reuse of those batteries in 2nd life applications would undoubtedly have positive impacts on climate change.

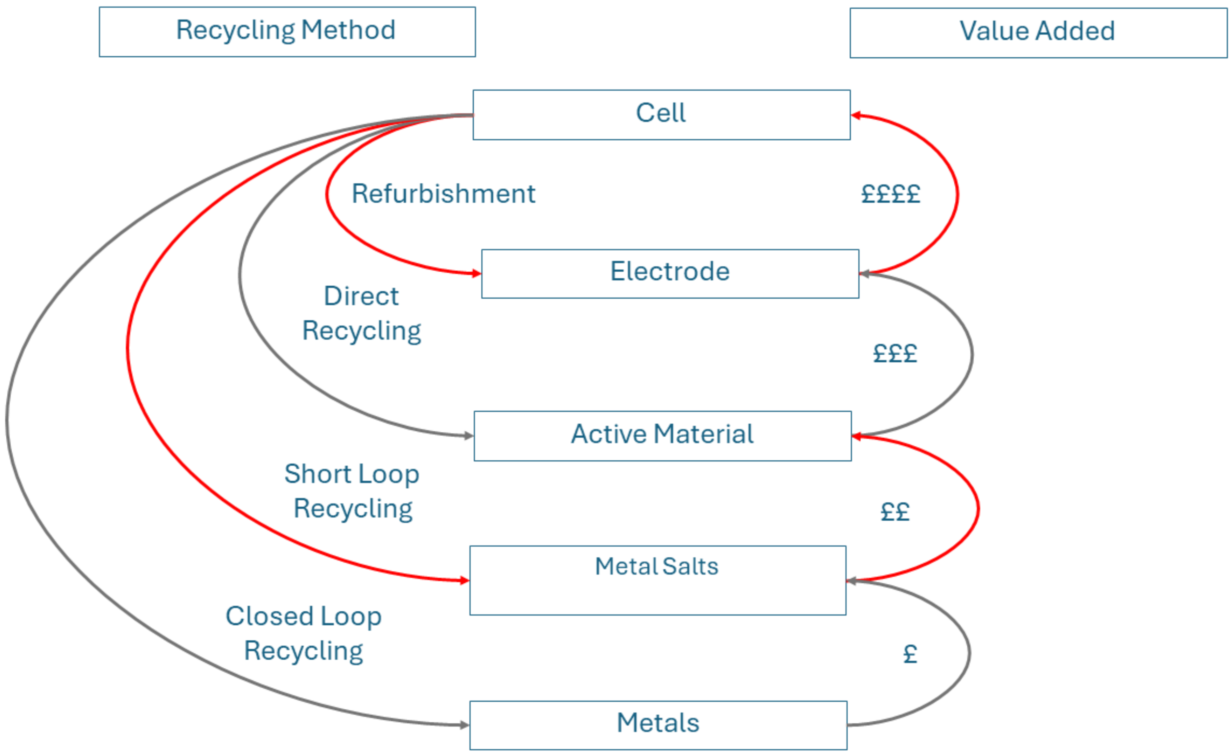


Figure 2.1: Waste management hierarchy. The left-hand side has the umbrella term for a family of processes. The right-hand side has the relative value added each time a material is processed up the ladder.

2.1.1 Direct Recycling

The cathode comprises mixed metal oxides, of which some elements (particularly Co, Ni, and Li) are critical, a rough breakdown of the contribution of each component to the cost of the cell can be seen in table 1.1. These can represent up to 65-70% of a cell's value [20]. Hence, this part of the cell has drawn the most attention regarding recycling. The most optimal strategy for any recycling method targeting the cathode is to recover the active material so it can be re-used with minimal post-treatment needed. These methods are known as direct recycling and can be defined as "the recovery, regeneration, and reuse of the cells' active material directly without breaking down its chemical structure" [28].

Direct recycling has been shown to work at a research lab scale for both end-of-life cells and manufacturing scrap. An upside to direct recycling is that unlike hydro- or pyro-metallurgical methods, where most of the value of the end product relies on the value of the Co or Ni, and this fluctuates significantly with time, the value of the outputs of direct recycling is tied to active material values that due to their nature are somewhat decoupled from the raw material prices. Also, direct recycling methodologies offer some future proofing with cell chemistries moving towards lower or entirely cobalt-free formulations. However, a downside is that the methods are likely to be chemistry-specific. That said, direct recycling promises to yield higher value outputs by maintaining the material's morphology [29]. Also, unlike pyro-metallurgy, other cell components can be recovered, providing further income streams for recyclers and potentially reducing environmental impact. An additional factor to consider is that the production of cathode materials embodies one of the most significant energy demands and environmental impacts, meaning that if the cathode material does not need to be re-manufactured, this can be mitigated [30] potentially reducing both costs and environmental impact. One of the challenges of direct recycling however is access to the cells as they are usually contained within modules and packs that are sealed or the cells are glued together and then welded to busbars. this requires the cells to be extracted as part of the recycling process adding cost and complexity [28, 31].

As battery innovation does not stand still and cells, in particular those used in the automotive sector, have extended life cycles, end-of-life waste streams are likely to contain a mix of cathode chemistries, meaning that some of the cathode active materials present may be already obsolete when they come to the recycling plant. This is a challenge to direct recycling methods as more simple techniques such as hydro- and pyro-metallurgy can overcome this by simply treating obsolete cells as ore enrichment, even if this comes at the cost of the product's value. By contrast, direct recycling relies on the ability of any pre-treatment steps to separate cathode materials into their various chemistries yielding sufficiently pure feedstock that can then be converted into not obsolete chemistries.

Many direct recycling processes of end-of-life cells shown at the lab scale rely on separating battery components; some require the active material to be entirely free of the binder and any other additives. This is challenging to demonstrate on a large scale. Therefore, a good halfway house for proof of concept of direct recycling may be the use of production scrap, which, with the ramp-up in electric vehicle manufacturing, is likely to constitute a not insignificant proportion of battery waste in the near future (this ranges from 5-30% scrap rate depending on if it is a top, middle or start-up manufacturer) [29]. Unlike end-of-life cells, manufacturing scrap should contain a well-defined chemistry and cathode formulation. Hence, it would not have to undergo cathode/cathode sorting.

Similarly, manufacturing scrap should comprise pristine or very lightly degraded cathode materials. After separation, it would require minimal processing before it can be remanufactured into a new cell. Therefore, manufacturing scrap shows a promising way for direct recycling to enter the recycling scene and be established as a proven technology, which would be aided by the fact that the battery industry is moving toward nickel-rich chemistries [30]. With production scrap being tightly controlled and well-defined this allows recycling technologies to be proven at a scale which should mean that once mixed stocks become available it is sorting and not the actual recycling process that will need to be optimised.

To help overcome the challenges above, methods of pre-sorting are being developed (particularly of shredded material) [32] and methods of post-treatment sorting of cathode material to aid separation based on the physical properties of the cathode materials, such as hydrophobicity and magnetism [33].

2.1.2 Hydro-metallurgy

Hydrometallurgy uses aqueous solutions to selectively leach metals from lithium-ion battery cathodes [34]. The initial step involves the disassembly of the cells. The use of a de-oxygenated atmosphere is beneficial when dismantling end-of-life cells [35] as this helps mitigate the risk of thermal runaway if any metallic lithium is present within the disassembled cell. Some pilot-scale hydro-metallurgical processes have been implemented, with reported recoveries of up to 75% of the active material [36]. Given the design complexity of LiBs, comminution serves as the primary stage, followed by a sequence of leaching and mechanical separation procedures. These include density-based separation, electromagnetic sorting, and hydrophobicity-driven techniques to separate the 'black mass', which is the mix of active material, binder and conductive additives [34]. Following this froth flotation can be used as a method to separate active material mixed metal oxides from carbon, however, sorting and categorising cells and their components may eliminate the need for this step [34]. Lv *et al.* has summarised the benefits and drawbacks of different recycling techniques [37]. After these pre-sorting and pre-treatment steps, if the black mass can undergo any processes that keep the morphology of the active material intact, it can be said that it has been directly recycled [34, 38]. the steps after disassembly or comminution for a hydro-metallurgical route are outlined below:

1. A solvent such as N-methyl pyrrolidinone (NMP) or dimethyl-sulphonate (DMSO) can dissolve the binder (this is binder specific and the solvent may change depending on the binder system) and detach the active material from the current collectors,
2. The active material is ground,
3. The active material undergoes physical separation,
4. Selected metals are leached from the active material,

5. The leachate is purified,
6. Precipitation, crystallisation, or electrowinning are used to obtain the final product.
7. The leaching step is key to the efficacy of hydro-metallurgical processing, its efficiency can depend on:
    (a) The reagents used and their concentration,
    (b) processing time and temperature,
    (c) The solid-to-liquid ratio,
    (d) Addition of any other reagents such as reducing agents.

The use of NMP as a solvent in the battery industry poses a number of health and safety risks as NMP carries a number of hazard phrases with it including causes skin irritation (H315), causes serious eye irritation (H319), may cause respiratory irritation (H335), may damage the unborn child (H360D).

Commonly, H_2SO_4, combined with H_2O_2, are used. Sulphuric acid is used as a very cost-effective lixiviant, mostly due to its availability as a by-product of various industrial processes such as flue gas capture and lead acid battery recycling. Alternative reagents that can be used include hydrochloric, nitric and phosphoric acids [34, 39]. One of the main issues with the use of H_2SO_4 and HCl is that they release toxic gases such as sulphur trioxide and chlorine, respectively, and these need to be removed from any emissions released into the atmosphere [40]. Organic acids such as oxalic, citric, maleic, and ascorbic acids have been investigated to try and mitigate this potential environmental impact [40, 41]. An alternative application of these will be discussed later. Concerning hydro-metallurgy, Zheng et al. [21]concluded that organic acids may not be sufficiently selective, since then a number of research groups have found methods to use organic acids to selectively leach transition metals or to aid in the selective recovery of critical materials from cathode materials [42–

45]. Another drawback of hydro-metallurgy is that generally, the process is very chemistry-specific and only effectively works on the chemistry that it has been designed for, meaning that each time a new chemistry family comes to market, the process must be redesigned [38, 46] (i.e. an NMC process can not necessarily be used on an LFP chemistry). Irrespective of this, a few companies, Umicore in particular have overcome these drawbacks by combining pyro- and hydro-metallurgy, but this has come at the detriment of the purity of the product [38].

2.1.3 Pyro-metallurgy

Pyrometallurgy is the most basic of the recycling methods and uses furnaces and a reducing agent to make alloys of Co, Cu, Fe and Ni [23, 34]. The main upsides of pyro- versus hydro-metallurgy are: it is chemistry agnostic, meaning it can deal with any cell, this removes the need for any pre-treatment (such as mechanical or pre-discharging), and it is safer as there is minimal handling of the cells [38]. Some major downsides, however, include high energy input leading to a potentially large carbon footprint and proportional cost (this can be offset by the use of renewable power sources for the heating of furnaces), loss of many lower value but still key elements such as Al, Li, and Mn to the slag, which is then sold on to the concrete industry, and the emission of harmful gases [21, 23, 34, 36, 38, 47]. Any plastics are also unrecoverable, and any pollutants must be removed before any gasses can be emitted [21, 34, 36]. Early in LiB recycling, pyro-metallurgy was preferred as it was already used in the extraction of Co and Ni so LiBs were simply used to enrich these ores. Also, capital had already been invested to understand the processes and build the plants. Umicore AG & Co. KG in Belgium and Sumitomo-Sony in Japan are the main users of pyrometallurgical processes.

Umicores process can in the best case scenario recover 50% by weight of material for reuse in batteries. Also, because Co is the target for recovery, this recycling process's economic viability relies on the chemistry's cobalt content and prices [48]. Umicore has invested £25 million into a plant in Belgium in anticipation of the growth of the EV market. Currently, they possess a capacity of 7 kilotonnes annually, equivalent to somewhere between 14,000 to 28,000 EV packs, although further explanations are planned. By comparison, the Finnish Akkuser and German Accurec have a processing capacity of 1 and 1.5-2 kilotonnes of batteries annually, respectively [23, 49]. With a global stock predicted to rise to 125 million EVs by 2030, it is obvious that recycling capacity must increase to meet demand [50, 51]. According to conservative estimates 8 million tonnes of EV batteries will come to an end-of-life (EoL) by 2030. At the current rate recycling will not be able to match that demand and virgin extraction is required to supplement the demand.

2.1.4 Cell Recycling Summary

The above section has discussed some current solutions and challenges in the lithium-ion recycling landscape. Some key points to take away are: 1) keeping material as close to its active cathode form as possible yields the most valuable and the least environmentally detrimental product. 2) Direct recycling, although challenging from a technological perspective, is probably the best method to achieve this. In the following results chapters, this work will tackle some small aspects of these challenges.

In Chapter 5, this work will look at organic acids. Can these be used for the direct recycling of cathode black mass? For this to be the case the recovered black mass must have greater capacity than the End-of-life cell (107.62 mAh/g as found by preliminary testing). Otherwise, can organic acids be used as a pre-treatment step for valorisation and hydrometallurgical treatments? For this, the leaching of metals must be controlled.

In Chapter 6, this work will investigate if DMSO can be used as a solvent to negate the binder in Nissan leaf generation 1 cathode to liberate black mass from the current collector, For this to be successful a separation of at least 85% black mass recovery to compare favourably with organic acid [52] and mineral acid delamination [53] and 99% black mass recovery to compare favourably with other reported solvent delamination techniques [54].

2.2 Cell Disassembly

Having understood some of the challenges facing recycling and the current state of the recycling landscape, we must now focus on how to get at the material in the cell before it can be recycled. This section will discuss the two most prevalent techniques: shredding and disassembly.

2.2.1 Shredding

The biggest challenges faced by recycling are the economic viability and environmental impact of processing. Shredding and post-processing are currently the cutting edge of lithium-ion battery recycling, aiming to address some of these challenges. Comminution aims to produce free-flowing and separated material to undergo physical separation to aid in post-processing [55]. Due to the free-flowing and separated nature of the material post comminution, further processing steps should be more cost-effective. To achieve comminution, various shredders and mills are used across multiple stages. The shredding, usually the first stage essentially opens the cell and produces a coarse fraction that can then be further refined, which is usually achieved by a high torque and low-speed shredder [56]. When shredding full cells, safety must be considered, and preventive measures such as engineering controls must be used to limit the probability of cells creating a thermal event; engineering controls such as extraction or high air flow would also be required to contain and control the VOCs, hydrogen fluoride, explosive atmospheres evolved due to cell opening, small particles that may be harmful if inhaled, and other hazardous components that are present in the cell [57]. During shredding, short circuits are inevitable. This creates the potential for rapid discharge and may produce excessive heat. If a flammable atmosphere (due to the evaporation of electrolyte components) is allowed to build up, this could lead to a fire. Engineering controls are used so that the risk of fire is limited. These include gas blankets to create an oxygen-poor atmosphere, water sprays can be used as heat sinks, or the use of rapid ventilation can also be used to not only control the temperature but also remove any flammable components in the atmosphere [58]. A pre-shredding discharge of the cells would limit the reliance on these engineering controls (e.g. extraction, atmospheric composition control, cooling, fire suppression etc) and potentially reclaim some energy for later use [56, 59]. After shredding, electrodes, casing, and separator can be segregated based on physical properties such as density, electronic conductivity, and susceptibility to magnetic fields.

A secondary comminution process can separate the current collectors from the black mass. This requires a much more high-speed process, such as hammer milling [58], the high-speed nature of this process enables the exploitation of the differences in how brittle the coating and the current collector are. This process takes advantage of the fact that the black mass is much more brittle than the metal current collector [60]; however, as discussed before, this may vary between production scrap and end-of-life materials [61]. An output that has undergone a two-step comminution process should be well-liberated, simplifying downstream processing and making recycling cost-efficient and effective. These two-step processes have shown great promise, achieving yields greater than 90% and low impurities (1.9 wt% Cu, 0.8 wt% Al and 0.3 wt% Fe) [36].

2.2.2 Disassembly

The secondary step of comminution will almost certainly produce fine particles that may cross-contaminate the finer fractions of active material. To combat this, disassembly may be used before comminution [62, 63]. Disassembly involves the manual or automatic separation of anodes, cathodes, separators, and packaging and is more time-consuming than shredding the cells. This produces material that is less contaminated by other components from the cell than shredding, and leads to simpler separation processes, however, it may be more hazardous than shredding, particularly if a human handler is required. It has been shown that even for relatively small batteries from non-pure battery electric vehicles, the process for pack-level disassembly can be very complex [64]. A Nissan Leaf Generation 1 pack can take up to 2 hours for a skilled engineer to manually disassemble without any module-level testing.

Careful disassembly of cells has several advantages over simply shredding cells. Previously, one study by Marshal *et al.* [62] focused on the disassembly of a single-cell type but did not consider other cell geometries or chemistries. Although manual disassembly appears to be the preferred method, at least from an input purity standpoint, the increased cost of human handling and the associated hazards are drawbacks; therefore, if disassembly is to be successful commercially, automation needs to play a large role in processing time, hazard mitigation, and cost [28, 65]. Disassembly does, however, fit well with the ideas of circular economy, enabling much purer feedstock for recycling processes.

2.2.3 Purity of mixed waste

As discussed in the shredding section, contamination is inevitable in this process. Some of the undesired contaminants are the separator and packaging material; the removal of this would prove beneficial to the energy requirement and cost of any further processing [66]. This can happen at two stages, either after all comminution has been completed or after the initial shredding that opens the cell and before the active material/current collector separation process [58]. Removing the packaging and separators before any further processing happens reduces the scale of the material needed to be processed, favorably impacting energy and cost demands. Removing the black mass from the current collector allows for the recovery of metallic Al and Cu, which are both costly and energy-intensive to recover from the salt form; this also prevents them from ending up in hydro-metallurgical processes where they add little value to the outputs [58]. Packaging can be easily removed by physical processes such as density or magnetic separation. Density separation can use either air or another fluid. During density separation, more dense materials, such as steel or Al from the packaging drop to the bottom, and less dense materials, such as the electrodes and separators, float to the top [36]. A secondary density separation step can then be performed on each of the fractions to separate Cu from Al (the dense fraction from the first process) and the current collector from the separators (the lighter fraction from the first process) [36]. Magnetic separation, although effective at removing metallic materials, is inefficient at separating plastics and electrodes [55]. Size separation can be used to separate black mass from the current collector after hammer milling, this can be achieved by a simple vibrating screen [55, 61]. Electrostatic separation exploits a difference in surface conductivity to segregate materials. This method has demonstrated efficacy in removing the non-conductive separator from the conductive electrode materials, resulting in both high grade and recovery percentage [67]. Higher recovery percentages with better purity reduce energy usage, resource demands, and cost and may also enable direct recycling processes, which require high purity and morphologically unaltered outputs [68].

2.2.4 Summary

This section has discussed the main issues facing the disassembly of cells, having compared shredding to disassembly. In these considerations, one thing becomes clear: recycling should not be an afterthought to the end of a cell's life, but cells (and higher architecture) should be designed from the ground up for recycling. This will be explored in Chapter 4.

The key contributions of Chapter 4 are the specific outline of how to disassemble the most common cells currently on the market, but most importantly the development of the methodology that allows for this to be applied to novel cell geometries as future markets change. There has been a considerable amount of work done on how to disassemble packs and modules [69–72] (a review of this is outside the scope of this work as it focuses on cell and smaller component treatment), to the knowledge of the author at the time of the writing of this thesis there is only one paper that concerns outlining a process of cell disassembly and to an extent the analysis of the components published by Marshal et al. [62]. This does not mean that this knowledge does not exist within research groups or industry, it is however not currently documented in an accessible manner for the range of the most common cells on the market. Furthermore, this chapter will also give recommendations on what would lead to a more recyclable and sustainable cell design.

2.3 Delamination

When a lithium-ion battery is no longer able to be used in a second-life application, such as grid storage or building backup systems, the preferable disposal route shifts towards recycling. Recycling presents a more environmentally sustainable alternative to manufacturing batteries from virgin materials, and this is important to create a more circular economy for critical materials contained within LiBs [73–76]. The main determinant of whether a battery is suitable for re-use in another application is if it has reached its knee point (this is the point at which the battery performance sharply and unpredictably drops off) [10, 77, 78], if it has, the battery is unsuitable in further applications due to the onset of sudden degradation and performance loss leading to an inability of the pack to provide sufficient power for vehicle acceleration and other drive cycle demands [10, 77, 78], necessitating its recycling.

Various component segregation methods exist for lithium-ion batteries, typically involving stabilization of the cell followed by shredding and comminution. Subsequent reclamation of components is achieved through a range of separation processes, including density, electrostatic, magnetic, or size separation techniques [55, 58, 67]. These processes yield several material waste streams, with the most valuable being the black mass. The most valuable black mass is the cathodic black mass, consisting of mixed transition metal oxides, lithium, binder, and conductive additives. The anodic black mass contains graphite and binders. These may be contaminated with some levels of current collectors: aluminium for the cathode and copper for the anode; however, depending on the separation, there may also be cross-contamination of the current collectors and black masses. The black mass can undergo further processing within a closed-loop recycling system. For this work, a black mass is defined as loose sheets of coating or free-flowing powder recovered from the coating on the current collector [52].

Closed-loop recycling generally has two approaches that can be classified as hydro-metallurgy or pyro-metallurgy. In hydro-metallurgy, metals are leached from the active material to form their respective salts. In contrast, pyro-metallurgy involves incinerating cells in a furnace to obtain a metal alloy containing the most valuable metals [79, 80]. However, these methods may face financial constraints, as they typically yield only a fraction of the original pack's value, estimated to be around 10%

As can be seen in Figure 2.2 different cathode chemistries have different inherent values

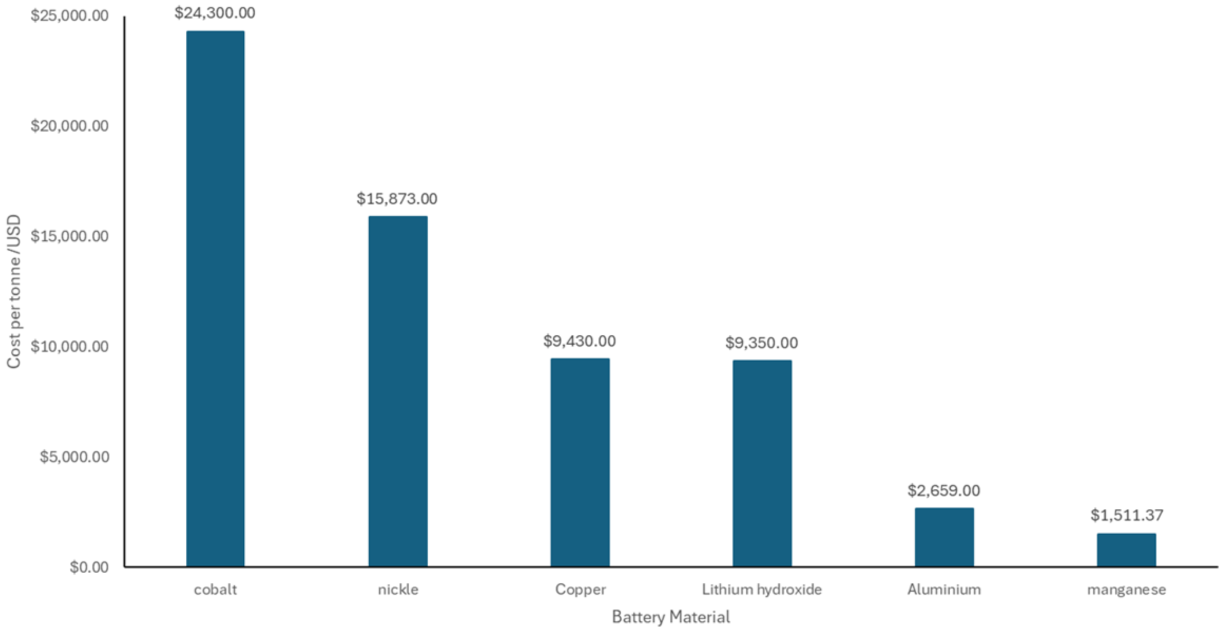


Figure 2.2: Cost of metals and lithium hydroxide used in the manufacturing of common lithium-ion batteries cathodes in USD per ton [81, 82]

due to their metal content. Therefore different recycling methodologies may be appropriate due to the cathode materials cost. Closed-loop recycling methods are particularly well-suited for processing lower-value materials, such as lithium manganese oxide (LMO), or for materials that are too degraded for direct recycling. This approach involves converting the black mass into its constituent components or metal salts for subsequent reuse. By contrast, short-loop or direct recycling methods focus on recovering the active material in its original active material form, requiring little post-processing. Material valorised by these methods is still in the morphology and crystal structure of the active material. Therefore, less energy is needed to re-manufacture the cathode, resulting in higher value retention. Short-loop recycling is particularly suitable for recycling materials generated as production scrap during manufacturing, enabling efficient resource utilisation and reducing environmental impact.

Closed-loop recycling has been demonstrated as a viable method [79, 80]. However, it has a drawback that prevents widespread adoption. These processes tend to be chemistry-specific to a family of cathode materials and hence may have to be modified and re-developed for the current and future chemistries [83]. The redevelopment of processes could be sped up if cells were marked with their chemistry as laid out in the EU directives discussed earlier, as this would go some way to prevent cross-contamination of chemistry-specific recycling processes. A more advanced and potentially economically viable approach would be direct recycling. However, there have been difficulties in demonstrating these methods [68]. Those methods that are presented in the literature use high volumes of water [84] and/or high temperatures [85, 86], which could be detrimental to the green credentials of these methods if the processes are not zero-waste, reusing the water, and the energy used for these processes is not from renewable sources. Acids have been demonstrated as leachants for transition metals [86–88]; however, these methods come with their drawbacks: some of these methods dissolve the Al current collector, which then, in turn, must be re-manufactured in an energy-intensive process. As all the metals are dissolved into solution in these processes, they require precipitation into salt, and some of these processes can be quite challenging [87].

Organic acids are a group of compounds that are defined by having at least one carboxylic acid group (-COOH). Although, from a cost and 'in terms of cost and supply chain efficiency, it may be easier to syphon off fractions of a percent of global mineral acid production (of an acid such as H_2SO_4). Organic acids, because of their variety offer several advantages over traditional mineral acids. Primarily, they can be derived from renewable and natural sources such as farm waste, biomass, or fermentation. This means these acids are considered renewable and potentially have a much lower environmental impact [89–91]. Additionally, unlike many mineral acids, organic acids can be biodegraded, reducing the environmental impact of their disposal [92–94].

Table 2.1 summarises current separation techniques that have been explored in the literature with some of their advantages and drawbacks. A few things to note in the drawback column of solvent dissolution and cryogenic grinding. Solvents dissolution can be expensive to set up due to the cost of solvents, however, this may be negated if the solvents are regenerated and continuously reused with only small amounts needed to top up the process once it is up and running. With regards to cryogenic grinding, this is only really viable as a small-scale batch process due to the cost and energy consumption of the production of liquid nitrogen and would only be viable for the most expensive or exotic materials.

Table 2.1: Summary of current separation techniques and their advantages and drawbacks.

Method	**Advantage**	**Disadvantage**
Mineral Acid Delamination [53]	Acids are easily manufactured, are low cost, and have a well-established supply chain.	Acids are single use. The acids carry hazards. Inefficient concerning waste. Direct recycling may not be possible due to leaching.
Organic Acid Delamination [89–91]	Acids can be considered a renewable resource as they can be manufactured via biological routes. Shared similarities with mineral acid delamination.	Acids have limited re-usability. Large quantities of waste may be generated.
Thermal Treatment [95, 96]	No high-tech methods required. Can be made chemistry-specific.	May emit harmful substances. The processes have to be tuned for specific cell chemistries.
Solvent Dissolution [54, 97, 98]	Solvent can be purified and used again. Effective rates of recovery.	Solvents may be toxic and flammable. Solvents can be costly.
Mechanical Grinding [55, 95, 99]	Well-understood methods, cost-effective.	Output requires post-processing due to contamination during comminution.
Cryogenic Grinding [55, 100, 101]	Efficient separation. The surface of the material remains unaffected.	High cost of liquid nitrogen. Difficult to scale. Depending on size, aluminium may be difficult to recover.

2.4 Electrodeposition Recovery

Electrodeposition, although not utilised in this work should be discussed as it could be a viable recovery method for dissolved metal ions in solution, depending on economic factors. Electrodeposition is a method that allows for the deposition of solid layers on a conductive material [102]. In lithium-ion batteries, a number of elements have the potential to be extracted using electrodeposition including Al, Fe, Li, Mn, Co, and Ni. Aluminium and iron are unlikely to be recovered by using electrodeposition though because of their relatively low value [103, 104]. The fact that lithium has a very negative equilibrium potential for the Li/Li^+ potential can lead to complications in the processing [105], however, the fact that lithium is a critical raw material within the EU may drive innovation in this area to overcome the complications. Electrodeposition of manganese is complicated and made more costly by the fact that it requires additives such as selenite and sulphite [106]. Nickel and cobalt make perhaps the most realistic candidates for electrodeposition recovery as they are both critical and strategic materials [107] in the EU and have ideal standard reduction potentials for electrodeposition. One drawback of using electrodeposition to extract cobalt and nickel from solution is the fact that their reduction potentials are very similar (-0.277 V and -0.257 V respectively), meaning that they commonly form a cobalt-nickel alloy that may need further purification [108].

2.5 Binder Negation

Both organic and mineral acids have been explored as a method to separate black mass from the current collector and have demonstrated good efficiency [52, 109–112]. However, these methods suffer from notable drawbacks. Firstly, the acids leach valuable transition metals from the black mass, meaning that without further post-processing, direct recycling (any process where recycled active material can be directly substituted for virgin material in the manufacturing of cells) is not possible [52, 111]. Secondly, in some reported studies, the acids dissolve the aluminium current collector entirely. This is undesirable as this could not only lead to aluminium contamination of re-manufactured active material, but the extraction of aluminium from solution is an energy-intensive process, so from an environmental and energy standpoint, it is considerably better to maintain the aluminium in its foil form and recycle through traditional metal recycling pathways [58].

Solvents have also been found to be useful in the recycling of EoL cells. The main advantages of using solvent instead of acids to separate black mass from the current collector are two-fold. First, they can be purified so that they can be used again and again. Second, they can dissolve the binder, making black mass purification simpler. However, the methods proposed in the literature so far have some drawbacks. N-methyl pyrrolidone (NMP), which is commonly used in the production of cells to dissolve the binder and therefore has been utilised in recycling, carries several health risks such as being teratogenic, skin and respiratory irritant, and flammable; it is also produced from oil refining by-products [113]. Glycerol, although less hazardous than NMP, is also produced from oil refining byproducts, and the method reported requires high temperatures ($T>100$ ℃) to be effective in short periods of time (15 minutes was found to be the optimal period for peeling and energy consumption) [114]. Cyrene has also been reported in the recycling of cathodes from LiBs. However, this requires high temperature ($T=100$ ℃) and an extended period (1 hour) to work [58, 109].

Dimethylsulfoxide (DMSO), the solvent proposed in this study, addresses several of the issues highlighted above. It is produced by treating dimethyl sulfide (a by-product of the Kraft process used to produce paper) [115]. This is environmentally better than using oil refining by-products, an LCA to quantify this benefit would be an interesting study, although it is out of the scope of this work. In its pure form, DMSO in its pure form is considered non-hazardous although when used with transition metals it has the ability to carry transition metals across the skin barrier, unlike NMP in its pure form, which carries a number of hazard warnings, and the methodology explored in this work shows that it is possible to achieve greater than 90% separation efficiency at low temperatures (T<100 ℃) and in short processing time (10-30 minutes). DMSO also has the potential to replace NMP in the manufacturing process. [116]

2.6 Achieving Direct Recycling

Although not directly investigated in this work it is important to look at what happens to black mass once it is separated from the current, and how direct recycling may occur. Sita et al. use LiOH at 700 °C for 15 hours in the air [117], so the output of a process similar to that as presented in Chapter 6 would be a good candidate for direct recycling here. Xu et al. [118] directly recycle LFP in an aqueous medium, they use NMP as the solvent to harvest the end-of-life LFP powders from the current collector. As the process uses NMP as the solvent DMSO could be a good alternative at the harvesting stage to provide powdered black mass for the re-lithiation step. Sloop et al. [68] had to perform first a hydrothermal reaction to separate cathode material from the current collector and then separate use froth floating followed by another heating step to 800 °C before producing recycled material. This process uses a lot of energy and can only be done as a batch process.

2.7 Research Questions

1. How do we disassemble lithium-ion cells so that all the components of these cells can be quantified?

2. What are the principles of safe disassembly of lithium-ion cells, and can these be applied

to any cell or electrochemical storage device?

3. What does the ideal cell from a recycling perspective look like?

   (a) *This work provides a safe, quantitative method for the disassembly of prismatic, cylindrical, and pouch cells. This methodology is then briefly applied to a cylindrical supper capacitor. finally in the discussion a commentary on what the ideal cell for recycling is provided.*

4. What is the possibility of using organic acids to directly recycle mixed active material cathodes from end-of-life battery electric vehicles?

   (a) *This work shows that organic acids may be used to separate and valorise cathode material, it also shows that due to the leaching of metals, it is not possible to do direct recycling with materials recovered using this method.*

5. Can DMSO be used in the process of separating cathodic black mass from current collector foil?

   (a) *This work shows that DMSO can be used to separate cathodic black mass from current collector foils with an efficiency of up to 95% at 60℃ in 10 minutes, depending on the feedstock of the process.*

Understanding a part of the current recycling landscape is important before assessing novel recycling and valorisation methodologies. It is essential to understand what is in a lithium-ion cell and characterise it to measure the success of a recycling process.

Chapter Three

Methods

Some of the methods presented in this section have already been published as part of Zorin, A., Song, T., Gastol, D. & Kendrick, E. Acid-Assisted Separation of Cathodic Material from Spent Electric Vehicle Batteries for Recycling. en. Metals 13, 1276. issn: 2075-4701. https://www.mdpi.com/2075-4701/13/7/1276 (2024) (July 2023).

In this section, will take a deep dive into the methods utilised in this work for analysis, and data generation. These include basic electrochemistry, x-ray diffraction, scanning electron microscopy and energy dispersive x-ray spectroscopy, and inductively coupled plasma optical emission spectroscopy.

3.1 Disassembly process flow

There are several common factors for any cell disassembly; the first is the cell's discharge. The cell, regardless of the format and chemistry, is discharged; this can be performed through various routes [119]. The cell's discharge first ensures the lithium is removed from the anode and inserted into the cathode. This can help recover lithium in future processes and enable a more direct recycling approach. In addition, the energy is removed, which means any thermal runaway event will be reduced in energy or eliminated. If there are enough cells, for example, in a module or pack, then the energy can be recovered. However, this is likely too time-consuming and costly for single cells. The energy can be discharged through a resistor or a brine solution. Certain salts will prevent any corrosion at the terminals. However, NaCl brines should be avoided, as these will corrode the terminals and allow water to penetrate the cell - which will complicate any future separation [120, 121]. The precise voltages should be determined by the cell chemistry [122]. However, if discharged via a resistor or salt solution, the self-discharge will occur until an equilibrium voltage is reached, which may not be desirable as this could cause copper dissolution and contamination of materials.

The disassembly and quantification consist of several steps. These are illustrated in the flow sheet in Figure 3.1. The flow chart starts with a full cell of which the outer dimensions, mass, and open cell voltage are measured. The cell is then disassembled and components are washed in IPA to remove contaminates such as the electrolyte salt. They are then dried in a bespoke heated vacuum chamber at 70 °C and 100 mbar. After this, physical characteristics such as the size and mass of the individual components are recorded. Then, samples were taken for more in-depth analysis, such as XRD, ICP-OES, and SEM-EDX, to determine the elemental and material composition of the components. The disassembled cell can be either stored under a vacuum (for a prolonged time to degradation as $LiPF_6$ has been removed) or used in further studies like those outlined in Chapter 5 and 6.

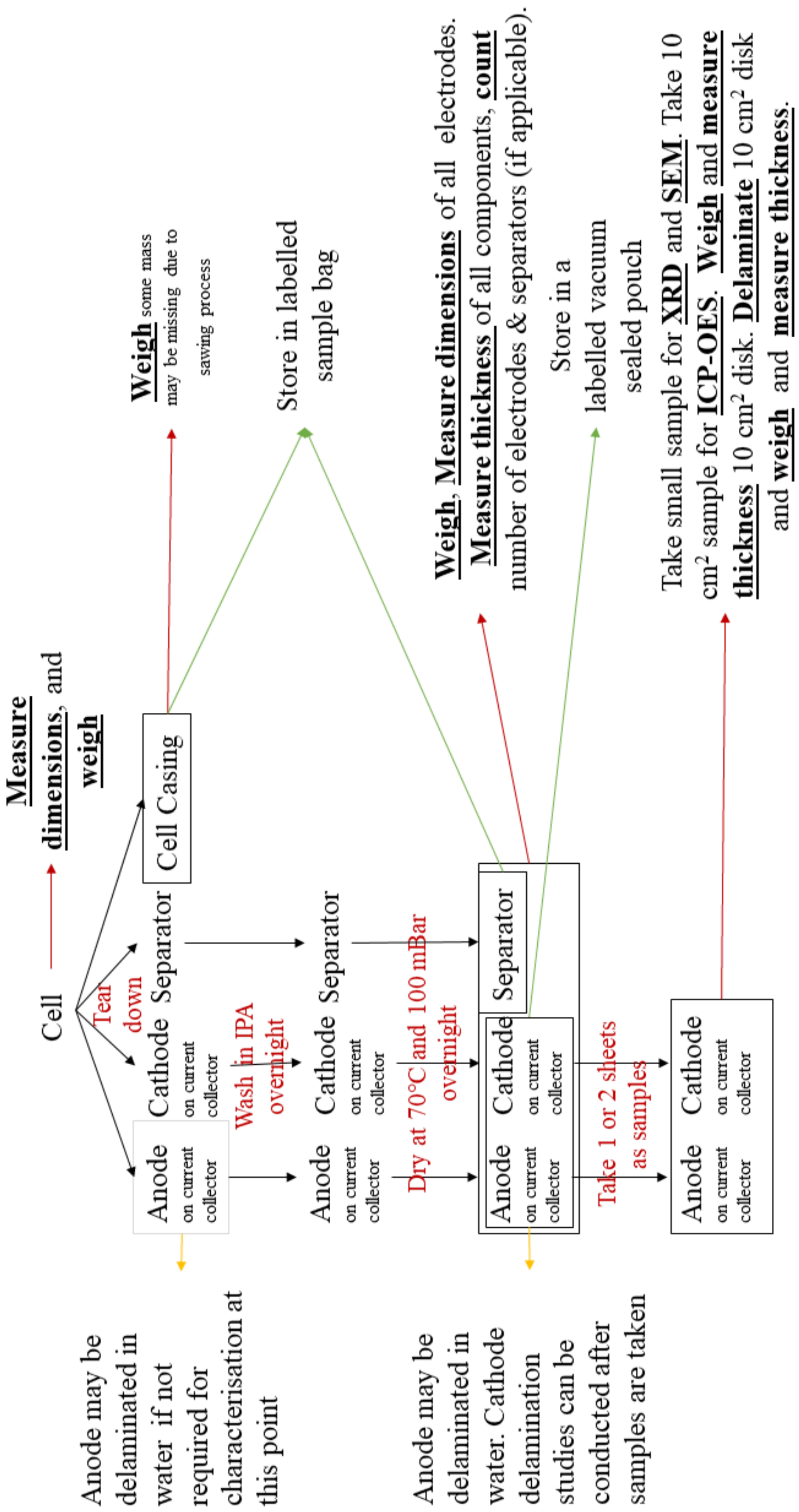


Figure 3.1: Process flow chart of the disassembly and characterisation steps for an electrochemical cell.

3.2 X-Ray Diffraction

X-ray diffraction is a powerful tool to understand crystal structure; it not only gives us an idea of crystal structure but it can also be used to understand the crystal composition and derive phase purity. Firstly before exploring the experimental setup, some of the basics of crystal structure should be explored.

3.2.1 Unit Cells

The most basic unit of a crystal is known as the unit cell and it is defined as the smallest and simplest repeatable unit [123]. All crystals are made up of repeating unit cells with a long-range order.

To be able to describe a unit cell 6 parameters and their measurements in the 3 dimensions must be known; these are commonly defined as a, b, and c and are given in Ångtsroms where 1 Å is 1×10^{-10}m. The angles between them must also be known, these are normally labelled as α, β, γ, and are given in degrees (°) this is illustrated in Figure 3.2 below.

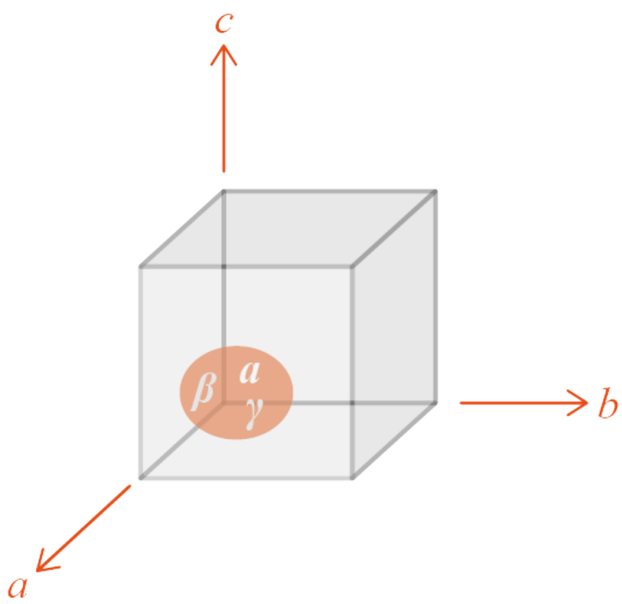


Figure 3.2: Schematic of unit cell parameters [124].

Expanding from the unit cell to create an extended crystal structure via translational symmetry, a crystal lattice can be inferred from the recurring units of the unit cells. This results in a spatial arrangement characterized by consistent and uniformly oriented surroundings, quantified by individual lattice points as illustrated in Figure 3.3.

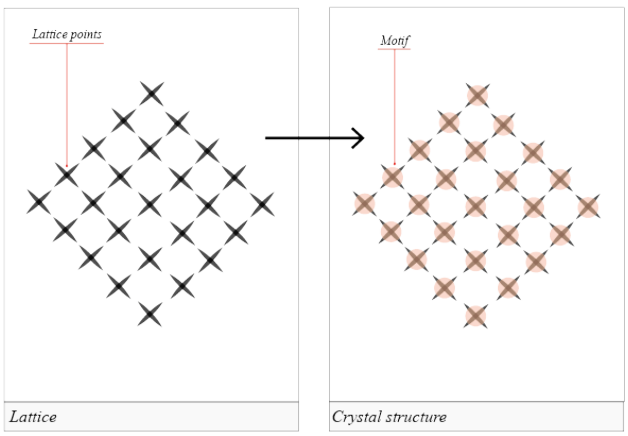


Figure 3.3: Using motifs to construct a crystal structure from lattice points [124].

3.2.2 Practical aspects of X-Ray Diffraction

X-rays produced within laboratory settings are generated by directing a beam of electrons accelerated to approximately 30-40 kV towards a metal target, such as copper (Cu). When these collide with the metal target, the incident electrons possess sufficient energy to ionize the inner electrons within the metal's 1s shell, commonly referred to as the K shell. This ionization process creates an electron vacancy within the inner shell. Subsequently, an electron from outer shells (e.g., 2p or 3p) transitions to fill this vacancy, emitting X-rays in the process. Because of the discrete nature of energy levels, the transition energies possess fixed values, resulting in a characteristic spectrum for the element. Because of this, only a very specific number of monochromatic wavelengths can be produced. It must be noted the resulting spectrum includes a broad spectrum of white radiation, generated from high-energy electrons decelerating or being halted upon collision with the metal target. The energy lost during these collisions is converted into electromagnetic radiation.

Turning our attention back to the characteristic wavelengths, the transition of the 2p electron to fill the 1s vacancy can be referred to as $K\alpha$, with a Cu target the wavelength of $K\alpha$ is 1.5418 Å, the transition of 3p to the 1s shell vacancy is commonly referred to as the $K\beta$ with a Cu target the wavelength of $K\beta$ is 1.3922 Å. The $K\alpha$ peak is of considerably higher intensity and therefore this means that the $K\alpha$ line is much more prominent in X-ray diffraction patterns.

To selectively filter out the unwanted $K\beta$ radiation and generate monochromatic wavelengths for experimental purposes, a filter is employed to eliminate undesirable wavelengths. For instance, when using Cu radiation, a sheet of Ni foil is commonly utilized. The higher energy of the $K\beta$ emission can efficiently ionize the nickel 1s shell (1.488 Å), enabling effective absorption of the undesirable $K\beta$ radiation (along with white radiation). In contrast, the $K\alpha$ radiation remains unaffected by the nickel filter, as depicted in Figure 3.4. It is worth noting that the atomic number of the element used in the filter is typically one or two less than that of the target material.

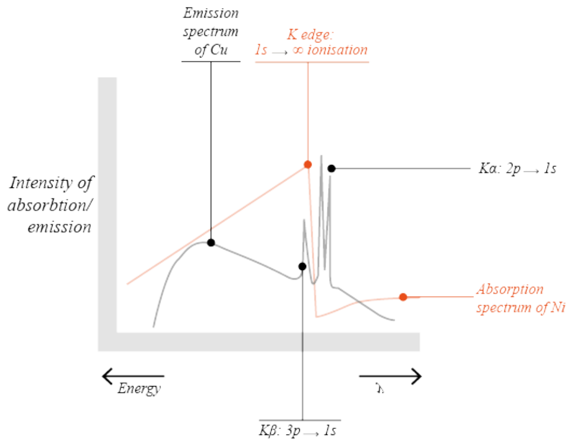


Figure 3.4: A diagram illustrating the emission of $K\alpha$ and $K\beta$ radiation, accompanied by the incorporation of a nickel (Ni) filter to remove the $K\beta$ radiation [124].

Another way to filter $K\beta$, is to use a crystal monochromator. This is done by carefully orientating the crystal monochromator in a specific way to allow for only $K\alpha$ radiation to be diffracted.

3.2.3 Bragg's Law

The Bragg approach views crystals as composed of distinct layers. Within these layers, atoms reflect incident radiation at an angle equal to the angle of incidence. Radiation may penetrate deeper into the crystal, subsequently reflecting off successive planes. Bragg's law is derived from the scenario where two rays of incident radiation are reflected. The second beam interacting with plane B must travel a further distance, denoted as XYZ, for both rays to remain in phase. This additional distance corresponds to an integral number of wavelengths, n, ensuring constructive interference and preventing destructive interference. This concept is illustrated schematically in Figure 3.5.

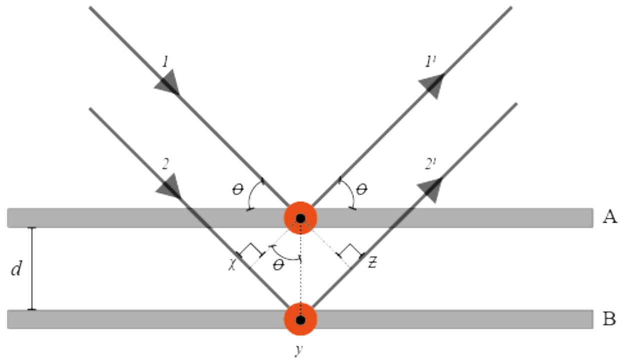


Figure 3.5: Illustration of the underlying principle behind Bragg's law, The reflection of incidence rays from two planes at different depths [124].

From this, the Bragg equation can be derived, leading to the expression shown in Equation 3.1. 2θ values obtained from experiment and using the Bragg equation enable the determination of d-spacing values, which, in turn, can be used to calculate unit cell parameters. The distinctive XRD pattern obtained serves as a fingerprint indicative of the composition of a specific crystal system.

$$n\lambda = 2d_{hkl}sin\theta_{hkl} \tag{3.1}$$

3.2.4 Powder X-Ray diffraction

X-ray diffraction and Bragg's law can not only be applied to single crystals but also to poly-crystalline powders, comprising of many small crystals commonly referred to as crystallites, arranged randomly relative to each other, can generate patterns using powder X-ray diffraction (PXRD). With a multitude of crystallites in the sample, there's a high likelihood that some planes within the material will meet the Bragg angle criteria for diffraction upon irradiation. The diffracted beams are detectable through a movable detector, and the data are collected using a computer.

From each lattice plane, diffracted radiation forms the surface of a cone due to the powder's random orientation, without constraints on Bragg's law regarding the angular orientation of crystallites. As the diffraction cones intersect with the detector, a trace of the corresponding intensity is recorded as a function of 2θ. this is illustrated in Figure 3.6

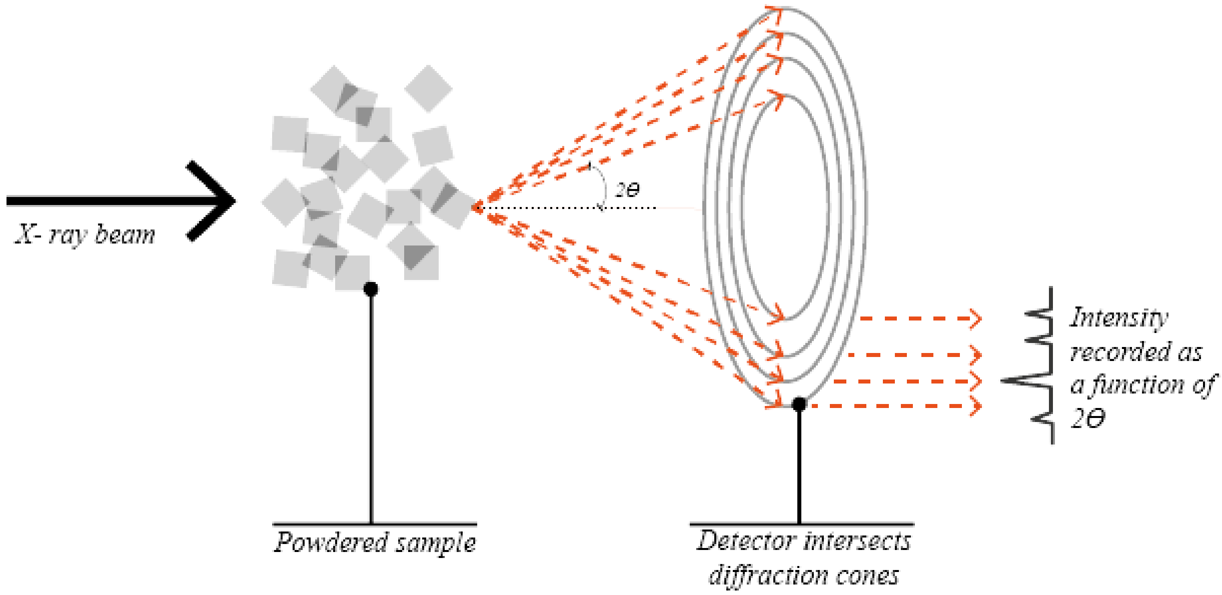


Figure 3.6: Schematic of a PXRD system, with incoming X-ray beam passing through a polycrystalline sample, being diffracted and forming a cone that is then detected and converted into a pattern [124].

3.2.5 Sample preparation and experimental conditions

Powder samples in this work, consisted of black mass and cathode active materials, were loaded into a sample cell, and levelled off using a glass slide ensuring there was enough powder to fill the whole cell. For samples that were coated onto a current collector a sample large enough to fill the sample holder was cut from the bulk. This was then mounted in the sample holder using plastic putty and a glass slide was used to ensure that the sample was both flat and level with the sample holder. In this work, XRD will be used predominantly in

Chapter 5. It will be used to determine if any changes have occurred to the crystal structure of a mixed-phase active material due to transition metal ion loss which changes the lattice parameters of the crystal.

Powder X-ray diffraction (PXRD) was performed using a Proto AXRD Bench-top diffractometer with a Cu K-alpha source 2θ from 10–80° with a step size of 0.01°, a dwell time of 11 s, and a $\lambda = 1.540593$ Å.

3.3 Scanning Electron Microscopy

Scanning electron microscopy is a powerful tool in the arsenal for characterisation and providing surface information. When combined with energy-dispersive X-ray spectroscopy it can also help visualise the distribution of elements.

When using this method, electrons from an electron gun are accelerated to an energy of between 5-50 kV and focused onto a small area of the sample (50 - 500 Å), capable of penetrating up to 1 µm deep. A schematic of the set-up of a scanning electron microscope (SEM) is depicted in Figure 3.7.

The SEM can operate in two modes: secondary electron (SE) imaging and back-scattered electron (BSE) imaging. These modes provide different insights into the material due to their differing energies, resulting in different electron escape depths. Secondary electron imaging captures inelastic scattering and can visualize the surface with a shallower depth, owing to lower energy. Back-scattered electrons can escape from deeper regions of the sample without further secondary collisions after the initial interaction. Electrons that penetrate beyond this depth are unable to escape due to energy loss to lattice vibrations. The penetration depths of both SE and BSE are illustrated in Figure 3.8.

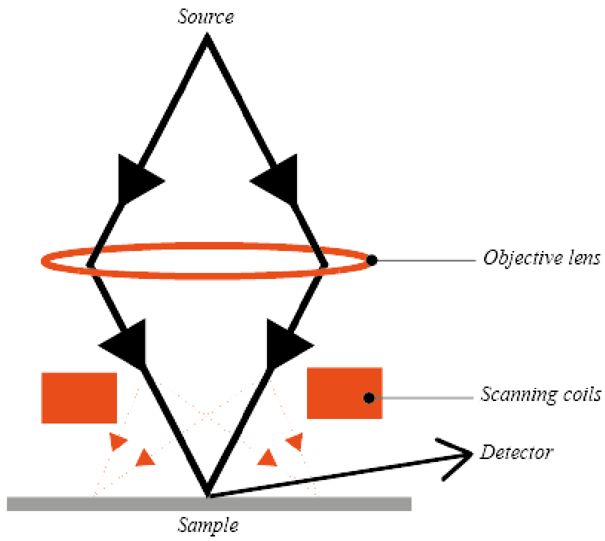


Figure 3.7: Reflection of radiation from different planes within a crystal, the main principle behind Bragg's law [124]

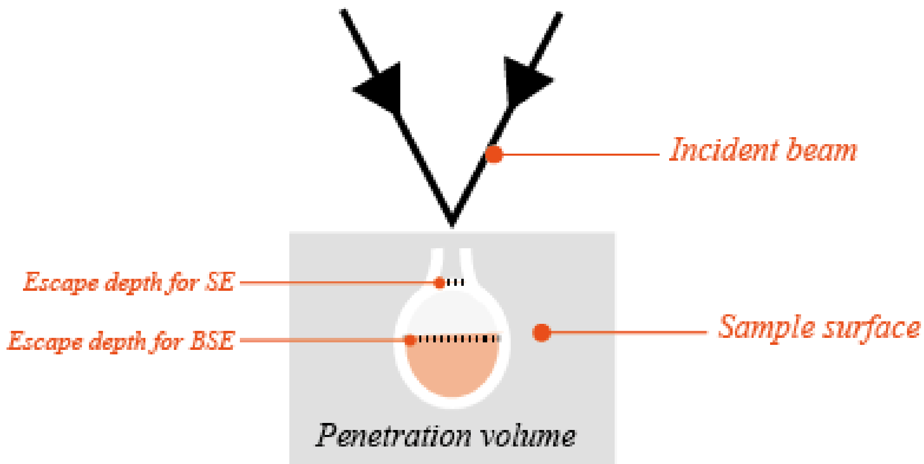


Figure 3.8: Schematic showing the relative penetration and escape depths of secondary electron and back-scatter electron imaging in scanning electrode microscopy [124].

3.3.1 Energy-dispersive X-Ray Spectroscopy (EDX/EDS)

Energy-dispersive X-ray spectroscopy (EDS), also known as EDX, facilitates qualitative analysis of elements present in a sample. By bombarding the surface with electrons in SEM, X-ray emission spectra are produced, allowing the determination of the elements' presence based on detected emitted energies/wavelengths. While quantitative analysis is possible, this requires extensive calibration, and this was not conducted in this thesis.

3.3.2 Sample Preparation

SEM-EDX was performed using a Hitachi 3030+ tabletop SEM (Hitachi, Japan). Samples were mounted on stubs using carbon stick dots with two samples per stub. Images were taken using 15 KeV using the EDX mode. EDX was performed using Aztek One (Oxford Instruments, UK).

3.4 Inductively Coupled Plasma - Optical Emission Spectroscopy

Inductively coupled plasma optical emission spectroscopy (ICP-OES) is a powerful analytical technique widely used for elemental analysis in many fields, ranging from environmental monitoring to materials science. It is particularly good at detecting metals in liquid samples.

3.4.1 Microwave Digestion

Microwave digestion is a technique to improve the dissolution of samples into solution. In this work, aqua regia is used (a 3:1 mixture of hydrochloric to nitric acid), a fresh batch is made for every digestion; then a small sample is placed into a polytetrafluoroethylene (PTFE) digest tube. This is sealed with a lid and placed into a carousel that is then placed into the microwave digestor. The carousel is spun to ensure all tubes are exposed to microwave radiation equally. Due to the fact that the tubes are sealed, there is a build-up of pressure within the tube, which allows for elevated temperatures beyond the boiling point of aqua regia at standard pressure. These elevated temperatures and pressures allow for the complete digestion of the metallic components into the acidic solution.

3.4.2 Instrumentation

The core components of an ICP-OES system are an ICP torch, a nebulizer, an optical spectrometer, and a detector. The ICP torch serves as the primary ionization source, generating a high-temperature plasma (> 6000 K) through the interaction of an inert gas (most commonly argon) with radio-frequency energy. The sample is introduced as an aerosol using a nebulizer, carried by the same inert gas as is used in the plasma torch, an illustration of a Meinhard nebuliser can be seen in Figure 3.9.

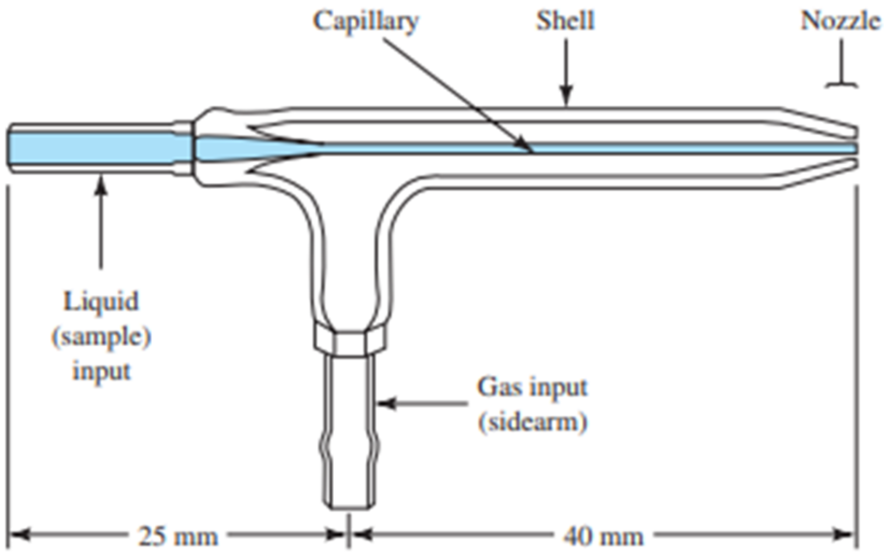


Figure 3.9: Diagram of a Meinhard nebuliser. Gas flows around a jacket which surrounds the capillary, due to Bernoulli's principle this creates a drop in pressure around the tip aerosolising the sample [125].

The optical spectrometer disperses emitted radiation into its constituent wavelengths using a diffraction grating or prism, allowing for the detection of specific spectral lines which are characteristic of each element. A photo-detector then quantifies the intensity of emitted radiation at these wavelengths. Using a calibration curve of known concentrations of the desired elements allows for elemental quantification.

The ICP-OES equipment used in this work has two sets of optical spectrometers and detectors. One is set axially to the torch and can detect low-intensity wavelength. The other is set radially to the torch and can detect high-intensity wavelengths (as illustrated in Figure 3.10.

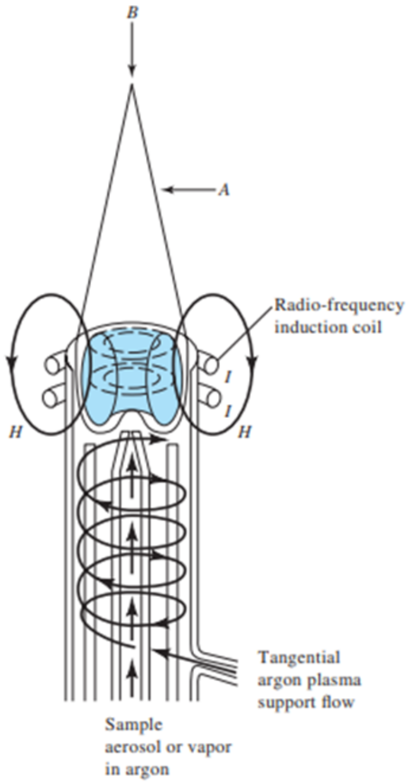


Figure 3.10: ICP torch A shows the direction of the radial detector. B shows the direction of the axial detector [126].

Upon introduction into the plasma, atoms in the analyte solution become excited by the high temperatures present in the plasma; as they travel up the torch, the outer shell electrons relax back down to the ground state hence emitting a characteristic wavelength of light that the detectors can detect.

Multiple elements can be detected at once in a single sample; however, when selecting the wavelengths to be detected care must be taken to ensure that the wavelengths that are selected are not interfered with by other species present in the sample. It is also important to ensure that the standards with which the ICP-OES has been calibrated contain all the elements that are being analysed across an appropriate range. In this work elements that were analysed included Li, Mn, Ni, Co, Fe, Cu, and P.

3.4.3 Sample preparation

Inductively coupled plasma optical emission spectroscopy (ICP-OES) analysis was conducted on the electrodes to analyse elemental composition. For powdered samples, 0.25 g of powder was digested. For coated samples (this is to mean actually coated electrodes i.e. current collector coated in slurry, dried, calendered, and usually constructed into a cell) 10 cm^2 discs were cut from each coating using a calibrated James Heal sample cutter (James Heal, Halifax, UK) these were cut down to weigh 0.25g or less. These samples were digested in a microwave digestor (Anton-Paar, Austria) for 45 minutes, maintaining an average temperature of 170 ℃. The digestion process was carried out in 10 mL of Aqua Regia. The digests were then filtered through quantitative filter paper and adjusted to a final volume of 250 mL using distilled water, ensuring that all glassware was thoroughly rinsed with distilled water beforehand. At this stage, three separate samples were collected.

A 10% dilution was prepared from the original 250 mL sample, and again, three samples were retained from this dilution step [127]. The same filtration and dilution procedures were also applied to the leachate solution.

ICP-OES analysis was performed using an Agilent 5110 ICP-OES instrument equipped with an argon plasma torch (Agilent Technologies, USA). Each sample underwent three repeated measurements on the instrument, with a rinse step carried out between each sample to prevent cross-contamination. Re-calibration of the calibration curves was conducted every 20 samples to ensure accuracy. The error tolerance was set at 20% (i.e if at re-calibration the value of the new standard point was more than 20% from the original the curve would have to be re-measured), and an r^2 value of 0.955 was targeted (this is the measure of how well the calibration curve fits the measured values, if this is less than 0.955, either the standard has to be discounted or re-made) for the calibration curves, the smaller the error tolerance the more accurate the measurement is and the close that r^2 is to 1 the more precise a measurement is. Both axial standards ranging from 0 to 15 ppm and radial standards ranging from 20 to 100 ppm were prepared for calibration purposes.

3.5 Electrochemical testing

3.5.1 Slurry making

Slurries were made using a Thinky ARE 250 centrifugal mixer (Thinky, USA). Initially, a binder comprising 8 wt.% PVDF Solef 5130 in NMP (1.44 g) was combined with 0.115 g of C65 conductive additive (Timcal) and 0.49 g of NMP solvent. The mixture underwent a two-step mixing process, starting at 500 rpm for 1 minute, followed by 2000 rpm for 5 minutes. Subsequently, 2 g of the recovered black mass was added to 0.49 g of NMP solvent, and the same mixing protocol was applied. The resulting mixture then underwent a degassing step at 2200 rpm for 3 minutes. The aim was to achieve a solid content of 50% [128].

3.5.2 Coating

The prepared slurries were applied onto aluminium foil using a draw-down coater (K Paint Applicator, RK Printcoat Instruments, UK) equipped with a doctor blade. Following coating, the samples were dried on a hot plate at 80 °C and then transferred to a 120 °C vacuum oven for 24 hours. After drying, the coating and foil underwent calendaring at 80 °C to achieve a porosity of 45% using an MTI calendar [128].

3.5.3 Cell Making

Capacity, rate, and cycle-life testing were conducted on Li-metal anode coin cells, as detailed later in this study [128]. The coat weight of the electrodes was adjusted to achieve an areal capacity of approximately 1 mAh/cm^2, based on a theoretical maximum capacity of 179.5 mAh/g. The dried coatings underwent processing in a dry room with a dew point not exceeding -45 °C.

The coatings were assembled into 2032 coin cells with a lithium metal counter electrode (15.0 mm, 150 µm thick), cathode electrode discs (14.8 mm in diameter), and a tri-layer 2025 separator (Celgard) (16.0 mm, 150 µm thick). These cells were then filled with 70 µm of PuriEL Battery Electrolyte (R&D281) from Soulbrain (MI, USA). The electrolyte composition comprised 1.0 M $LiPF_6$ in EC/EMC (ethylene carbonate/ethyl methyl carbonate) 3/7 (v/v) + 1% wt. VC (vinylene carbonate) electrolyte. Finally, the cells were sealed using a hydraulic crimper (MSK-110, MTI Corporation, USA).

3.5.4 Base testing and formation

In this study, galvanostatic testing served as the primary cycling methodology, involving the application of a constant current while monitoring the voltage response. All electrochemical testing was conducted using a Bio-Logic BCS 805 series cycler.

For 1st cycle capacities and formation, the following protocol was implemented: delithiation at 10 mA/g to 4.3 V vs. Li/Li^+, followed by lithiation to 3.0 V vs. Li/Li^+ at the same current density. This process was repeated for two consecutive cycles under these conditions. After formation, cycling was performed by applying a charge cycle at 80 mA/g and a discharge cycle at 320 mA/g, for a total of 50 cycles [128].

3.6 Material extraction

Material extraction will be discussed in depth in Chapter 4 where a methodology for the quantitative disassembly and assessment of various cell geometries is discussed. An important distinction between materials extracted to be noted is that between fresh and aged material which will be mentioned again in Chapter 6. Fresh material was used within 2 weeks of disassembly. To make the aged material, a cell was disassembled, in a fume cupboard, washed in propan-2-ol to remove electrolytes and dried at 70 °C at 100 mbar. It was then exposed to ambient conditions for a minimum of 4 weeks to undergo potential degradation and calendar ageing. This was done to simulate disassembly sites not being near one another and to investigate if calendar ageing of disassembled cells impacted the performance of the solvent-based recycling method.

Chapter Four

Quantitative Disassembly

4.1 Introduction

As stated previously, recycling of materials is a key to creating a circular economy in the electrification of the automotive sector, as this enables an industry where materials are kept within the sector and geographical region rather than using new primary feed-stocks and importing them from overseas. This is key not only from an environmental but also from the stability of supply [129–131] perspective. The recycling of bulk materials in this sector (such as glass, steel, and aluminium) is well established with high recycling yields [63]. However, more complex and composite materials like those found in lithium-ion batteries are much more difficult to recycle. Before these can be processed, we must understand the composition and design of the initial feedstock. If the design of the feedstock is understood, then the separation of the components and materials will be more targeted and designed into the recycling process, enabling greater recovery rates. Without that understanding, many valuable and critical materials may slip through the process as waste. This chapter attempts to lay out firstly a safe way to disassemble the three most common geometries available today and provide guidelines for developing safe methods for future novel cell geometries, then present a suite of analysis tools that allow the understanding of what is contained in the cell starting with a mass balance and going down to an elemental breakdown, finally, the chapter will provide a summary critique of design for disassembly and recycling and make suggestions for improvements. The cells used in this chapter (and in the case of the Nissan Leaf Generation 1 cells used throughout this work unless otherwise specified) have the following provenance; Nissan Leaf Generation 1 pouch cells are over-discharged production scrap; Calb prismatic cells are end-of-life cells that underwent over-discharge due to a failure in the battery management system; the LGM50 cells are end-of-life cells that have undergone research laboratory testing.

4.2 Methodology

4.2.1 Prismatic Cell Disassembly

The prismatic cell in this case was a Calb 130 Ah cell. This plastic-encased rectangular cell weighing 4.4 kg, and 17.8 cm by 27.4 cm by 6.9 cm (see table 4.1). Notable external features include large terminals on the top with large bolts around each and a safety vent that could be unscrewed with a hex wrench (Figure 4.8). From the purposes of quantitative disassembly, the main benefit of this type of cell design is that due to the rigidity of the packaging, internal voids within the cell can be reliably predicted for safe opening procedures. Also in this cell design the separator was of the z-fold configuration, meaning that disassembly could be easily automated as the separator can be simply wound around a spool and this would eject the electrodes in different directions [132, 133]. The rigidity of the casing provides challenges for quantitative disassembly, as either great force has to be applied to overcome friction forces to remove the stack if only one side is removed, or multiple sides have to be removed. This is similar to the cylindrical cell but far more difficult than the pouch cell.

The cell was weighed, and the external dimensions were measured, including the sizes and position of the external electrical contacts. At this point, any fastenings that may be holding the casing onto the internal stack (these may be large nuts etc) were removed. If necessary, incisions were made at diagonally opposite corners first, to drain any liquid electrolyte. Using a rotary cutting tool or a hand saw cuts were made around the perimeter of the top of the cell (along the red line in Figure 4.1), ensuring that the cuts go through the casing but did not cut the tab/tag connection; this is allowed by the air gap that is present at the top of this cell. These gaps can be identified either by x-ray tomography[134] or by having a cell that had undergone an event and could be easily seen into. Using compressed air, any debris from the cutting was removed and collected for weighing later. Gentle upward pressure was applied to the stack to check if it would slide out of the casing, if this was not the case one of the long sides was cut away and removed (Figure 4.2), as can be seen in the figure there is not a lot of space so it must be ensured that cutting is done in a controlled and precise manner. Once the stack was free of the casing, any fastenings between the tags and tabs were removed so that the tabs could be weighed separately from the electrodes. If the tags are welded onto the tabs, these will have to be cut. The stack was then disassembled into its constituents (anode, cathode, separator) and washed as described in the methods chapter (Chapter 3).

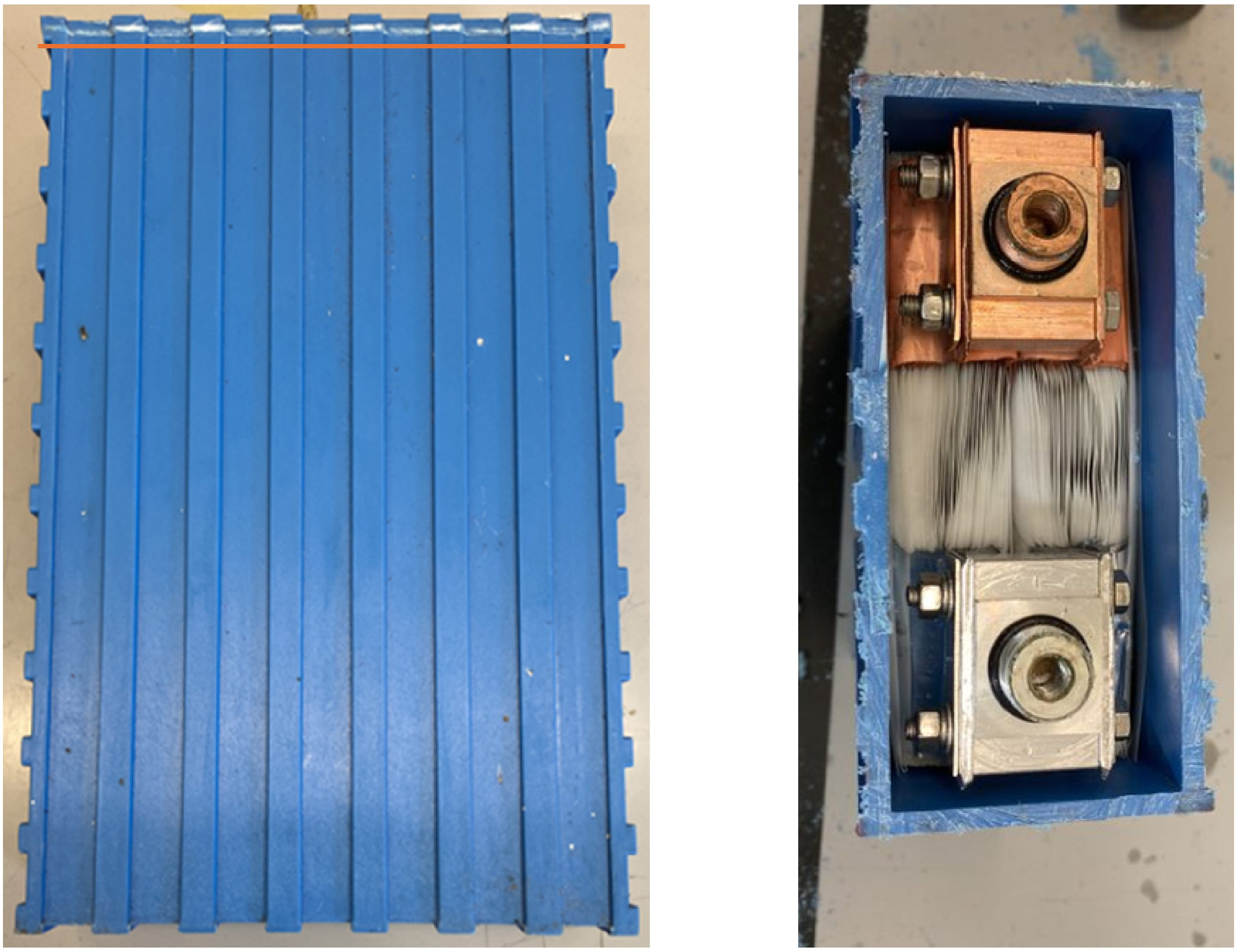


Figure 4.1: Opening cut for Calb prismatic cell shown in red with the top of the cell removed.

Figure 4.2: Long side cut (shown in red) of Calb cell and the side removed.

4.2.2 Pouch Cell Disassembly

The pouch cell comprises 4 main components: the outer packaging that is laminated aluminium, the cathode, the anode, and the separator. A sub-component that is attached to the outer packaging is the tabs, which are aluminium, and nickel-coated copper for the cathode and anode respectively. This construction remains true for pouch-type cells irrespective of size. For the purposes of quantitative disassembly, the benefit of this type of construction is that the pouch material is very simple and easy to cut with a blade, which means the removal of the stack is very simple when compared to the prismatic and cylindrical cells as there are no large friction forces involved. The downside of this cell format is that due to the flexibility of the pouch material, the electrode stack and the pouch can bend and deform, making standardisation difficult. Also, the fact that in this particular cell design, the separator is part of the heat-sealed edge, no method of cutting that generates high temperatures can be used as this would melt the separator, resealing the electrodes. Another disadvantage of this particular cell design is the stacked construction of the electrode-separator stack meaning that each layer has to be individually moved rather than easily automated as can be done with z-fold or jelly roll electrode stack construction [132]. Another challenge is the fact that each separator sheet is sealed into the heat seal of the pouch in 3 points along the two long sides and the bottom meaning that it is imperative to cut through all the separators before commencing further separation. As mentioned the drawbacks inherent to this particular pouch cell design can be overcome by using a z-fold or rolled stack that is not sealed into the pouch material edges. The choice of electrode stack geometry (i.e. z-fold or rolled) is dependent on the manufacturer.

The cell is weighed and the external dimensions of the cell are measured, including the sizes and position of the external electrical contacts. Using a ceramic scalpel, an incision is made along the edge of the pouch cell. The cut should be between the edge of the pouch cell, where the two sides of the pouch material meet, and the stack of electrodes inside the cell as shown in Figure 4.3 and Figure 4.4. The cuts should be made along three sides of the pouch, preferably both the long sides and the bottom (the short side without the tabs). To avoid short circuits and possible thermal runaway electrodes should not be cut. Fold open the outer layer of pouch material to reveal the electrode stack (Figure 4.5). A non-conductive ceramic scalpel can be used to cut the tops of the electrodes near the tabs below the welds, which reduces the chance of a short circuit occurring. The stack can now be disassembled into its constituents (anode, cathode, separator) and washed as described in the methods chapter (chapter 3). There are two predominant internal configurations for pouch cells: Stacked and z-fold [135]. Although these differ in the stack disassembly which will be discussed later, the initial opening of the cell remains the same.

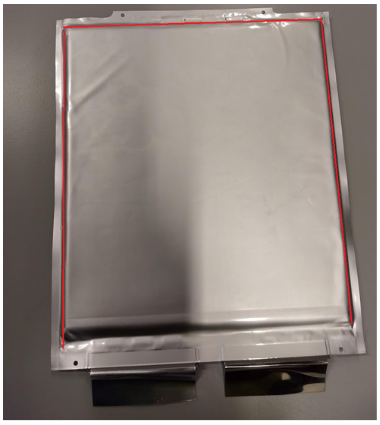


Figure 4.3: Image of where to make cuts (cut lines in red) on a Nissan generation 1 pouch cell.

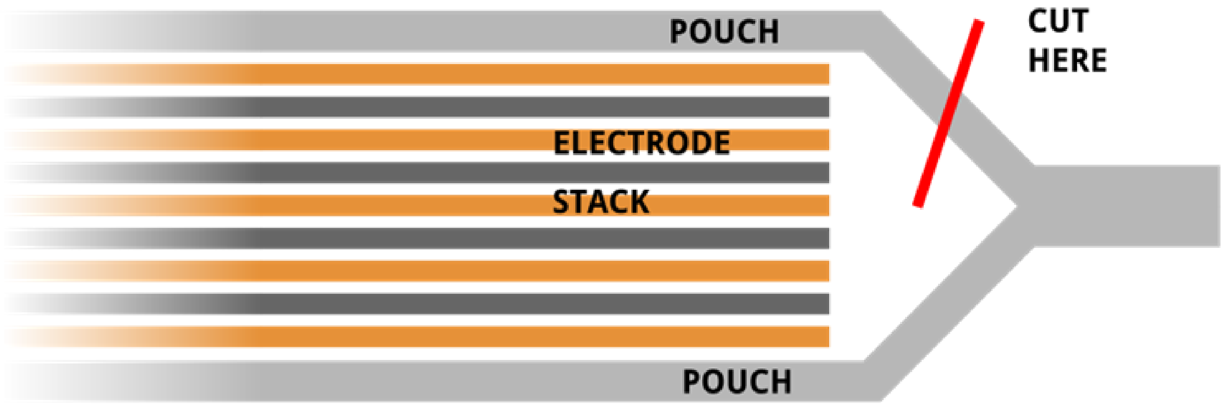


Figure 4.4: Schematic showing the void into which it is safe to cut, on a Nissan generation 1 cell.

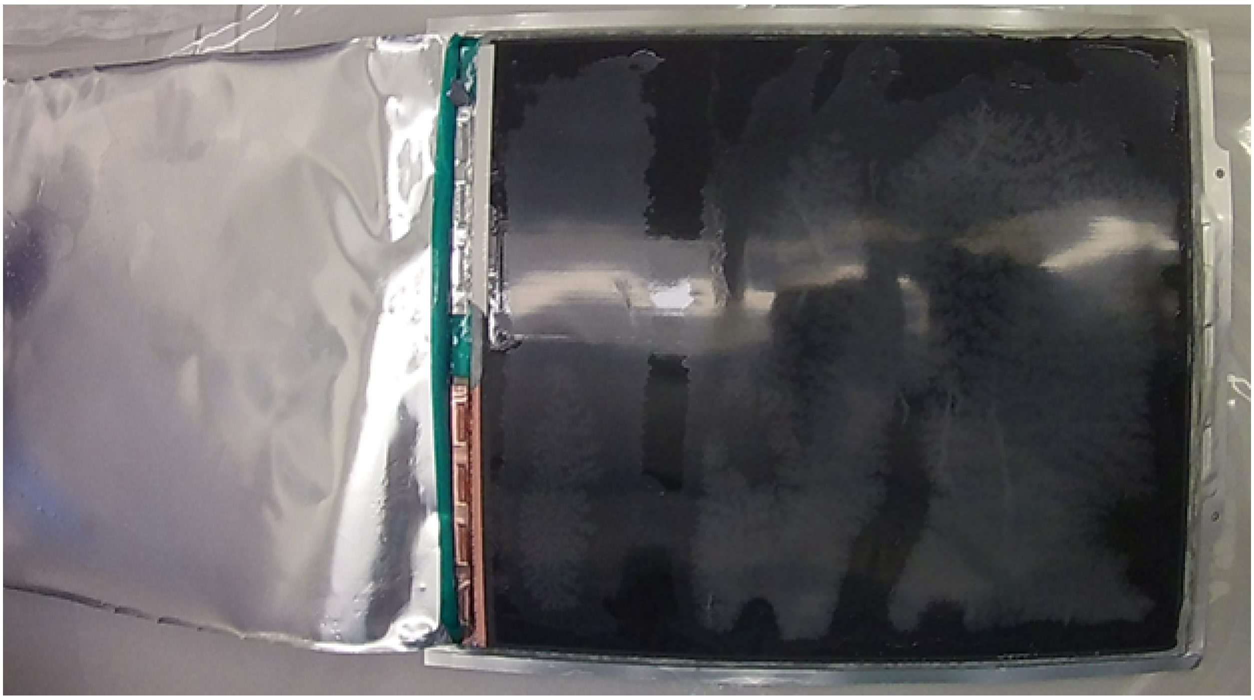
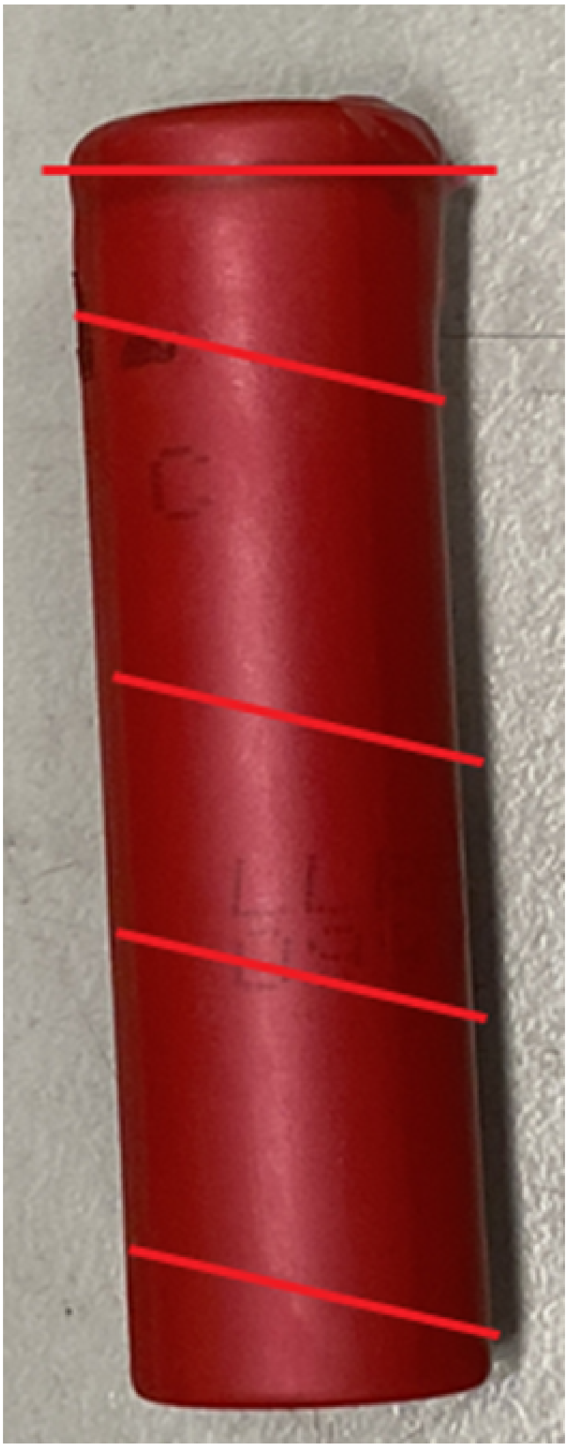


Figure 4.5: Image showing the cell after making incisions down 3 sides of the pouch and opening the top layer of pouch material to reveal the electrode stack. The green bar across the tabs/tags is insulating tape.

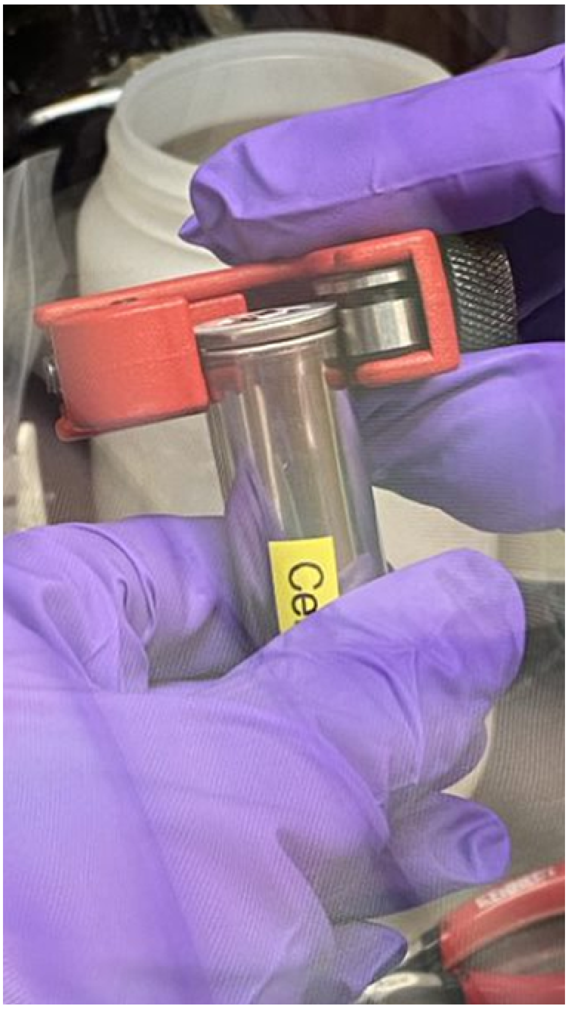
4.2.3 Cylindrical Cell Disassembly

The weight and the external dimensions of the cell are measured, including the sizes and position of the external electrical contacts. A pipe cutter is used to gently tighten into the divot around the top of the can and rotate through 360 $\circ$ as shown in Figure 4.6b. This process is repeated until the top of the can is released. This was attached by a small length of the current collector as can be seen in Figure 4.7, which is cut using ceramic scissors. Pliers were used to pry away the casing from the roll in a spiral pattern, as in the summary in Figure 4.6a until the jelly roll could be easily removed from any remaining can material. The jelly roll is attached to the bottom of the can by a small length of foil, this is cut using ceramic scissors or a ceramic scalpel. It is possible to count the number of winds from the jelly roll directly or from a photo. The anode, cathode, and separator are unwound and separated to be washed as described in the methods chapter (Chapter 3.

The main advantage of cylindrical cells is that they are energy-dense as they have very little dead space. This lack of dead space however causes a large problem for manual disassembly. There is only one place to make an incision at the safety valve end of the can to which the anode is attached. There is then very little space to peel away the can, and no space to make any secondary incisions to accelerate this process. This can be overcome with the use of an automated laser or blade to remove the can as these have much tighter tolerances, however, this may require some initial testing and manual disassembly to understand where cuts should be made. Another drawback of manual disassembly of cylindrical cells is due to the tight packaging of the jelly roll within the can, it is necessary to remove a large portion of the can before the jelly roll can be removed. The only benefit from a disassembly perspective of this format is that there is only one each of the cathode, anode, and separator, meaning that separation, once the jelly roll is out of the can, is trivial.



(a) Summary of cuts and peeling shown as red lines on the exterior of LGM50 cell.



(b) The cutting of the top of the can using a pipe-cutter.

Figure 4.6: Images of the disassembly of an LGM50 cell: a) a summary of the cuts that are to be made (the bulge at the top of the cell is due to shrink wrap plastic going over the top of the safety cap), b) The use of a pipe-cutter to cut away the top of the can.

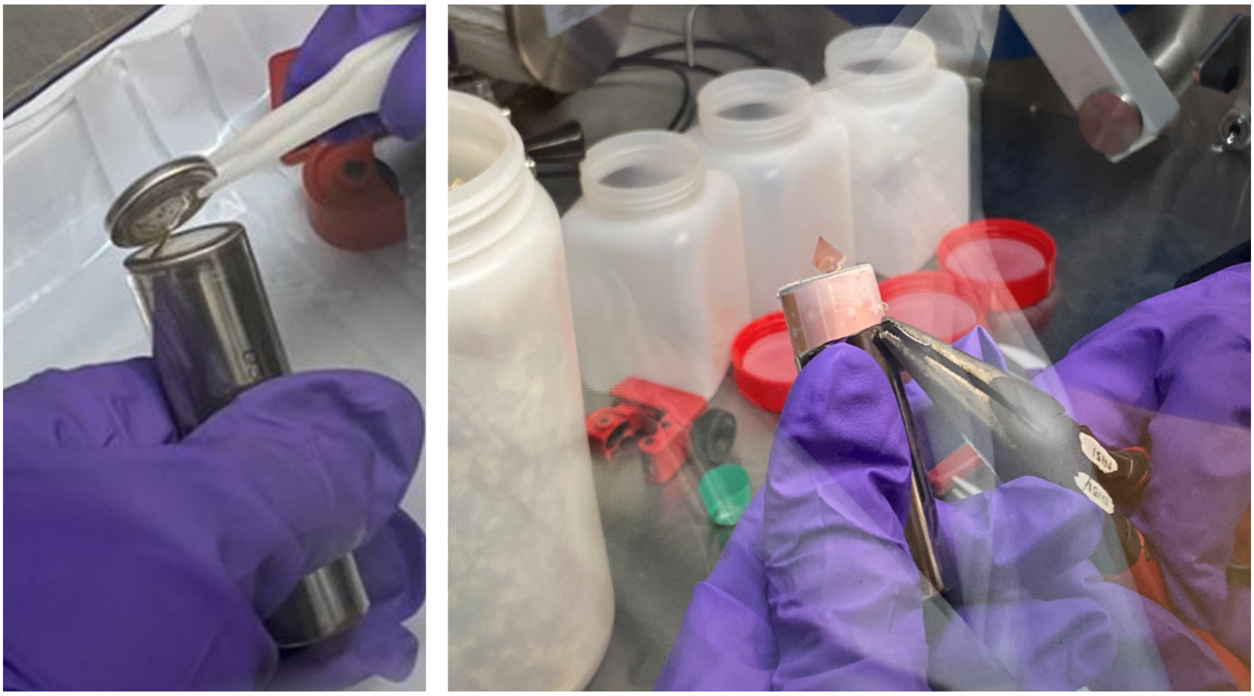


Figure 4.7: The removal of the top (safety valve) of the cell and spiral peeling away of the can from the jelly roll.

4.3 Results

In this section, the components and construction of the three cells being investigated will be analysed and discussed. We will look at the construction of the cells and their makeup.

4.3.1 Physical Characterisation and Schematics

Prismatic: Calb 130 Ah

The Calb 130 Ah cell is a prismatic cell encased in hard plastic. Both terminals are at the top of the cell and are held in place with nuts. The anode terminal is solid copper and the cathode terminal is solid aluminium. Between the terminals, there is a safety pressure release valve consisting internally of a plastic plate and spring. There are two electrode stacks with a z-fold plastic separator. The cathode is of an LFP chemistry and the anode is graphite. The binder systems on both the anode and cathode are unknown. It is not PVDF as it does not delaminate with the application of NMP and there is no F signal present in the EDX. In addition, it appears to be water-based; the application of water to the coating does not dissolve it, however, it does seem to flake off (rather than dissolve) when exfoliated. The lack of a Na signal in the EDX suggests this is not CMC. Overall the EDX mapping, at least with respect to binder identification is inconclusive.

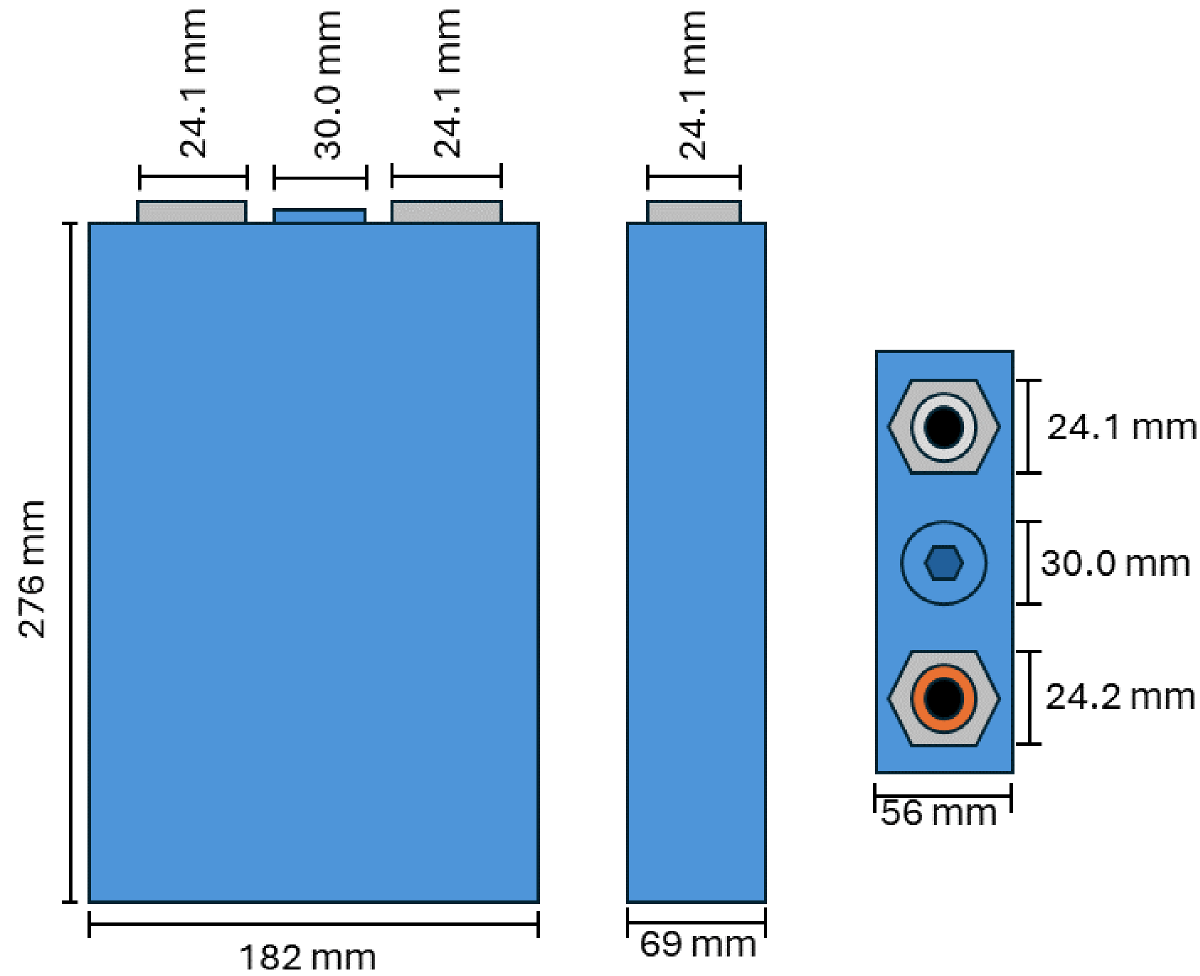


Figure 4.8: Exterior schematic showing the Calb 130 Ah cell.

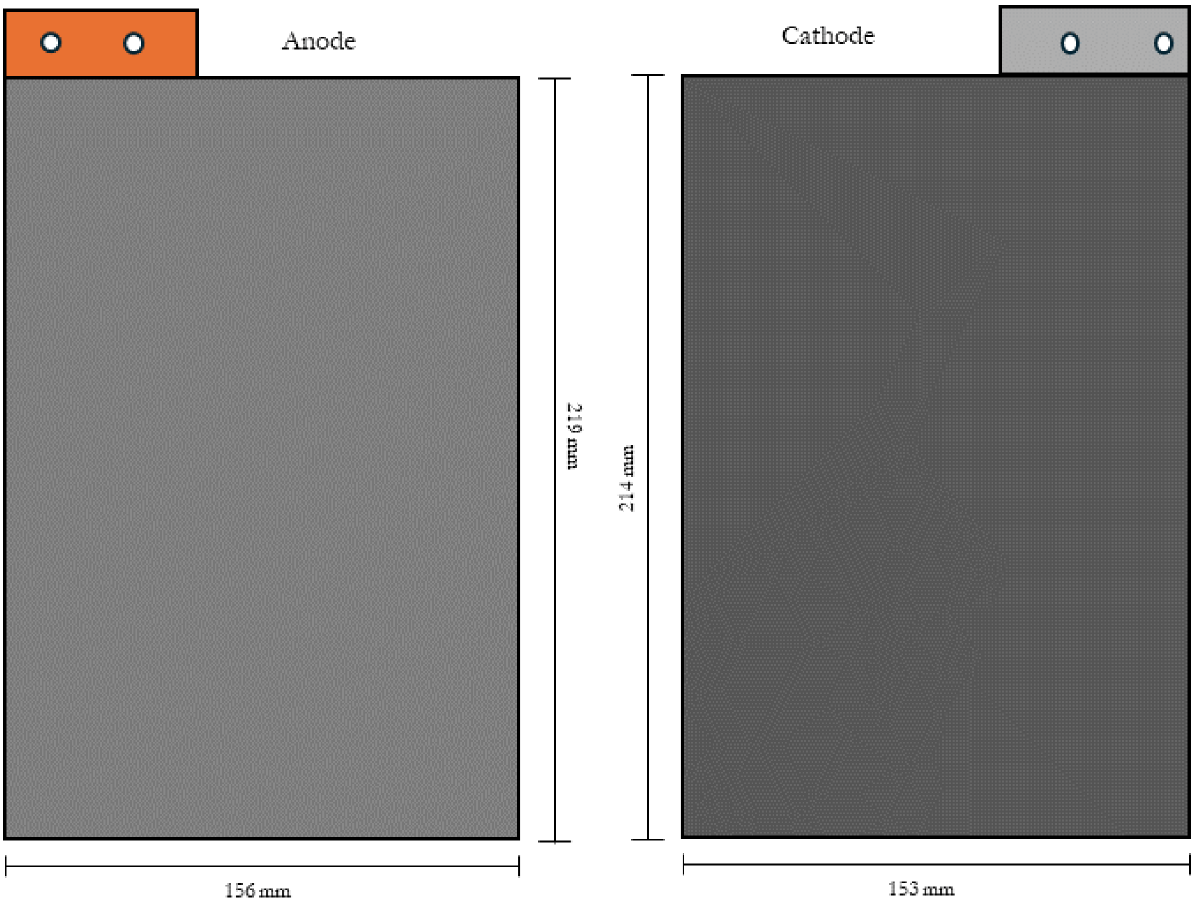


Figure 4.9: Schematic of Calb 130 Ah cell electrodes.

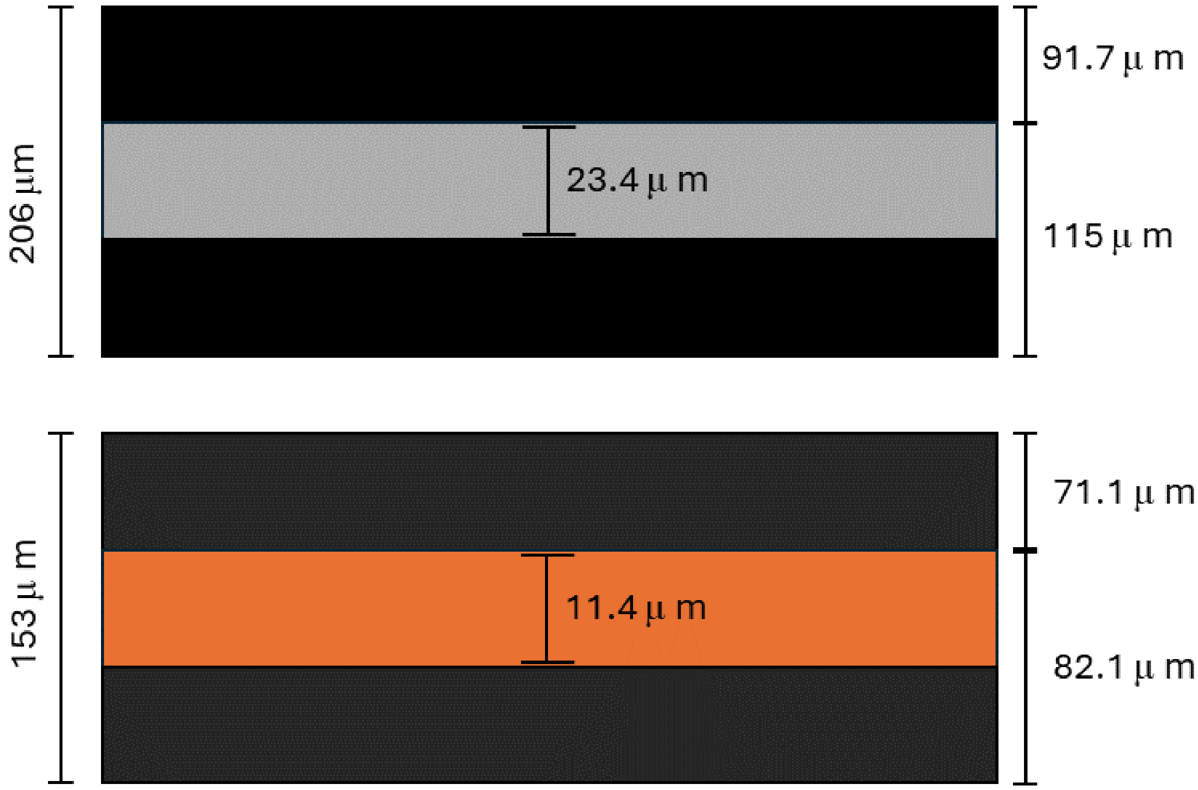


Figure 4.10: Schematic showing the cross-section of the Calb 130 Ah cell electrodes.

Table 4.1: Physical cell Parameters of Calb 130 Ah cell.

Parameter	**Value**
Manufacturer	Calb
Type	Prismatic
Serial Number	cel1000309lfp130h201205050001
Outer width (mm)	178
Outer Length (mm)	274
Tab positions	top of case, see schematics
Tab Width Cathode (mm)	15.5
Tab Length Cathode (mm)	
Tab Width Anode (mm)	15.3
Tab Length Anode (mm)	0
Weld Length Cathode (mm)	N/A as tabs and tags are bolted and not welded together
Weld width Cathode (mm)	N/A as tabs and tags are bolted and not welded together
Weld positions Cathode	N/A screws
Weld Length Anode (mm)	N/A as tabs and tags are bolted and not welded together
Weld Width Anode (mm)	N/A as tabs and tags are bolted and not welded together
Weld positions Anode	N/A screws
Cell Mass (g)	4368.25
Stack / Jelly Roll Mass (g)	2786.05
Electrolyte Mass (g)	784.35
Packaging Mass (g)	797.85

Table 4.2: Physical Electrode Parameters of Calb 130 Ah Cell.

Parameter	**Anode**	**Cathode**	**Separator**
Electrode Foil Length (mm)	219	214	N/A
Electrode Foil width (mm)	156	153	N/A
Electrode coating width (mm)	156	153	239
Electrode coating Length (mm)	219	214	1484.5
Electrode Surface Area (m^2)	0.34	0.33	3.55
Foil Surface Area (m^2)	2.06	2.20	3.55
Number of Electrodes	102	98	2
Current Collector thickness (μm)	11.4	23.4	18.71
Current Collector Type	Cu	Al	Plastic
Double-sided thickness + current collector (μm)	153.1	206.8	N/A
Electrode coating Thickness single side (μm)	82.5	115.1	N/A
Coating Density (gsm)	116.6	203	N/A
Electrode stack Mass (g)	1060.6	1530.95	197.25

Pouch: Nissan Generation 1

The Nissan generation 1 cell is a soft-shelled pouch cell constructed out of plastic laminated aluminium [136] outer casing with a layer-by-layer stacked electrode. The edges of this laminate are heat-sealed with the separator pouches protruding out into the heat seal at three anchoring points along the two long sides and the bottom side. The anode is graphite with a PVDF binder, the cathode is a mixed active material cathode consisting of LMO:NCA also with a PVDF binder. The electrodes are attached to the tabs via tags that are ultrasonically welded at 3 points. A schematic of the cell is shown in Figure 4.11 alongside a cross-section schematic of the electrodes in Figure 4.12. All the parameters measured are summarised in tables 4.3 and 4.4

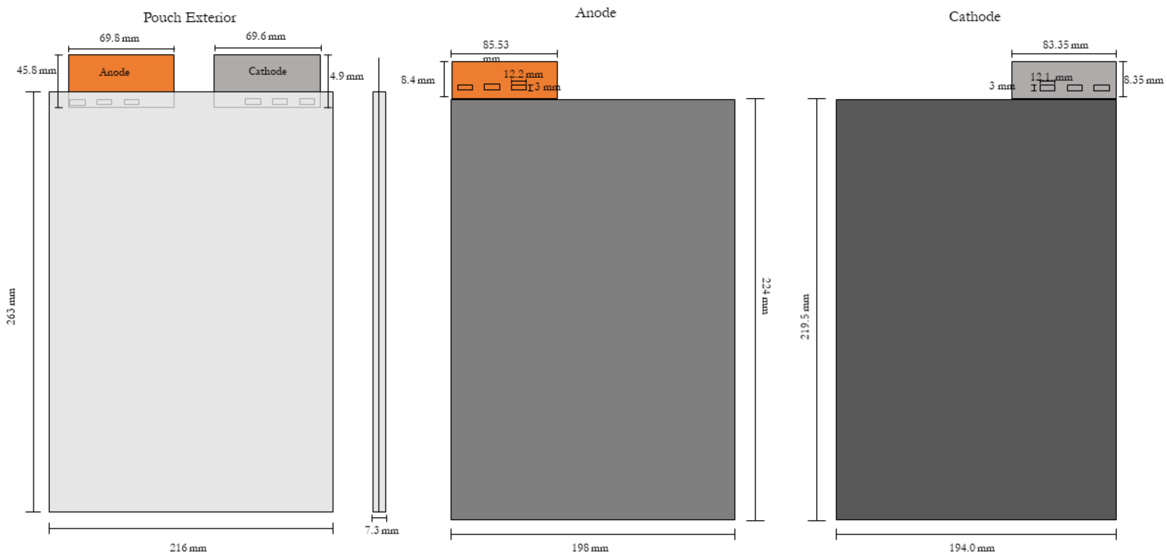


Figure 4.11: External pouch cell and full electrode schematic.

Figure 4.12: Schematic of electrode cross-section of Nissan generation 1.

Table 4.3: Physical cell Parameters of Nissan Generation 1 Cell.

Parameter	**Value**
Manufacturer	Nissan
Type	Pouch
Condition	Over discharged production scrap
Serial Number	E70-50 NBZG59JA0393
Outer width (mm)	215
Outer Length (mm)	256
Tab positions	Top edge of pouch
Tab Width Cathode (mm)	69.9
Tab Length Cathode (mm)	46
Tab Width Anode (mm)	69.7
Tab Length Anode (mm)	45.8
Weld Length Cathode (mm)	12.3
Weld width Cathode (mm)	3.6
Weld positions Cathode	long welds x 3 perpendicular to terminal/tag
Weld Length Anode (mm)	12.8
Weld Width Anode (mm)	2.8
Weld positions Anode	long welds x 3 perpendicular to terminal/tag
Cell Mass (g)	792.1
Stack / Jelly Roll Mass (g)	627.05
Electrolyte Mass (g)	128.15
Packaging Mass (g)	36.9

Table 4.4: Physical Electrode Parameters of Nissan Generation 1 Cell.

Parameter	**Anode**	**Cathode**	**Separator**
Electrode Foil Length (mm)*	same as coating	same as coating	N/A
Electrode Foil Width (mm)*	same as coating	same as coating	N/A
Electrode Coating Width (mm)	196.5	194	204
Electrode Coating Length (mm)	224.5	219.5	226
Electrode Surface Area (m^2)	0.44	0.43	0.46
Foil Surface Area (m^2)	0.57	0.54	0.46
Number of Sheets	18	17	34
Current Collector Thickness (μm)	11	20.57	16.71
Current Collector Type	Cu	Al	N/A
Double Sided Thickness + Current Collector (μm)	139.86	181.64	N/A
Electrode Coating Thickness Single Side (μm)	76.29	101.5	N/A
Coating Density (gsm)	96.1	234.5	N/A
Electrode Stack Mass (g)	222.3	386.3	18.5

*foil length and width are the same as the coating size in this cell

Cylindrical: LGM50

The LGM50 cell is a cylindrical 21700 cell, with a nominal 21 mm diameter, and 70 mm length. The casing consists of a steel can with a safety valve at one end. There is a plastic grommet that insulates the body of the can (which acts as the positive electrode terminal) making a connection with the safety valve which acts as the anode terminal. The can is wrapped in plastic insulation to prevent contact with anything conducting externally (see Figure 4.13. The jelly roll consists of a single anode, a cathode, and a separator tightly wound. The anode is a graphite SiOx mix with a water-based binder. The cathode is an NMC chemistry with PVDF as the binder. The electrodes are attached by a small thin tag ultrasonically welded to the can. The cell parameters are summarised in table 4.5 and electrode parameters in table 4.6.

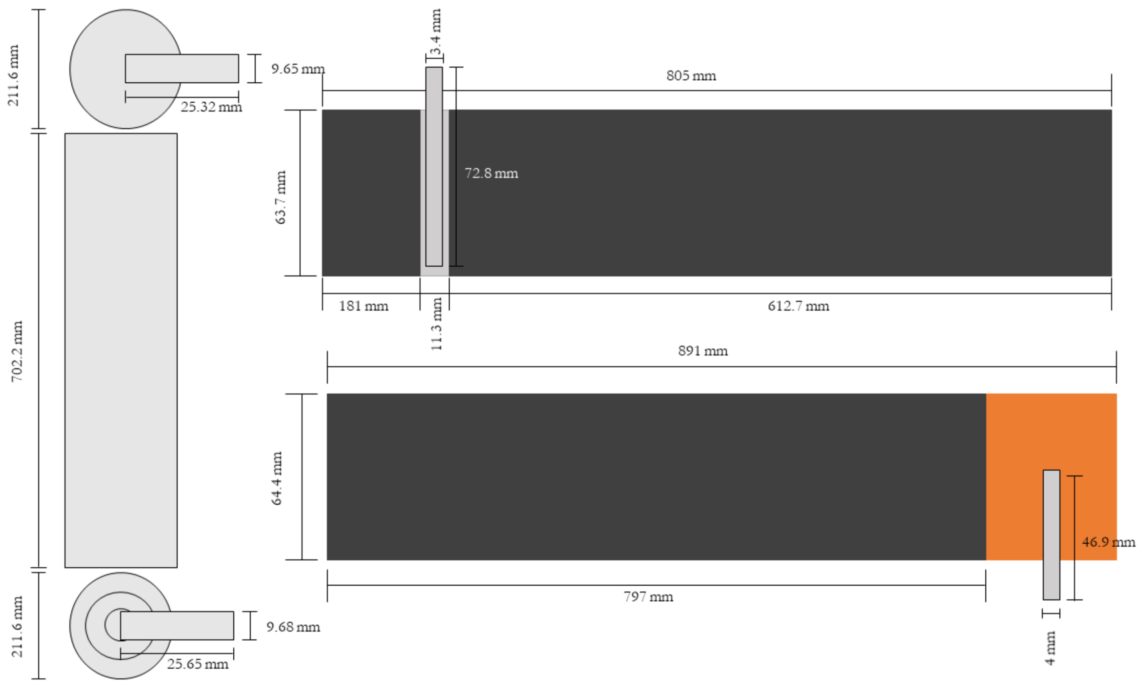


Figure 4.13: External can and Full electrode schematic

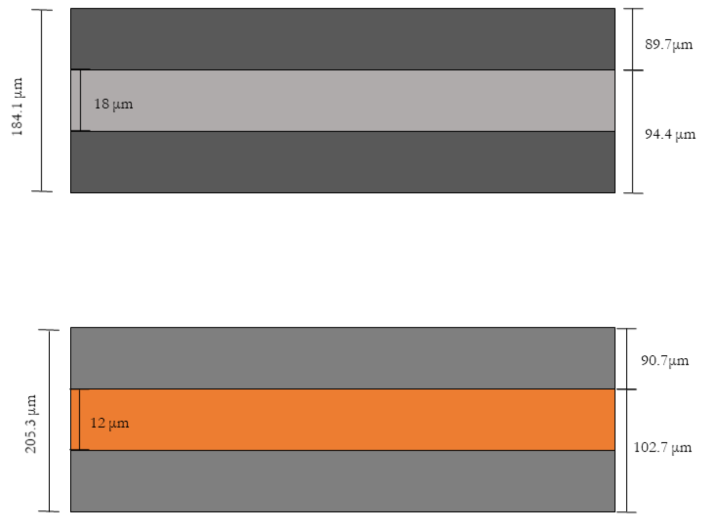


Figure 4.14: Schematic of electrode cross-section in LGM50 cell.

Table 4.5: Physical cell Parameters of LGM50 Cell.

Parameter	**Value**
Manufacturer	LG Chem
Type	M50
Serial Number	LGDBM502170Q340L061A3
Outer width (mm)	21.16
Outer Length (mm)	70.22
Tab positions	top and bottom of cell
Tab Width Cathode (mm)	9.65
Tab Length Cathode (mm)	25.32
Tab Width Anode (mm)	9.68
Tab Length Anode (mm)	25.65
Weld Length Cathode (mm)	N/A
Weld width Cathode (mm)	N/A
Weld positions Cathode	N/A
Weld Length Anode (mm)	N/A
Weld Width Anode (mm)	N/A
Weld positions Anode	N/A
Cell Mass (g)	70.133
Stack / Jelly Roll Mass (g)	52.03
Electrolyte Mass (g)	5.12
Packaging Mass (g)	12.983

Table 4.6: Physical Electrode Parameters of LGM50 Cell.

Parameter	Anode	Cathode	Separator
Electrode Foil Length (mm)	891	805	1937
Electrode Foil Width (mm)	64.4	63.7	65.1
Electrode Coating Width (mm)	64.4	63.7	N/A
Electrode Coating Length (mm)	797	793.7	N/A
Electrode Surface Area (m^2)	0.0513	0.0506	N/A
Foil Surface Area (m^2)	0.0574	0.0513	N/A
Number of Electrodes	1	1	1
Current Collector Thickness (μm)	12	18	10
Current Collector Type	Cu	Al	Plastic
Double Sided Thickness + Current Collector (μm)	205	184	N/A
Electrode Coating Thickness Single Side (μm)	102.7	94.4	N/A
Coating Density (gsm)	146.35	254.15	N/A
Electrode Stack Mass (g)	22.0662	28.4645	1.4982

4.3.2 Elemental Analysis, and Microscopy.

Figure 4.15 shows the results in PPM of the elements Al, Co, Cu, Li, Mn, and Ni that can be found in 10 different samples of cathode material taken from across a single Nissan generation 1 cell. The error bars represent the standard deviation present across 3 repeats from each sample. The table 4.7 shows the exact numbers shown in the graph.

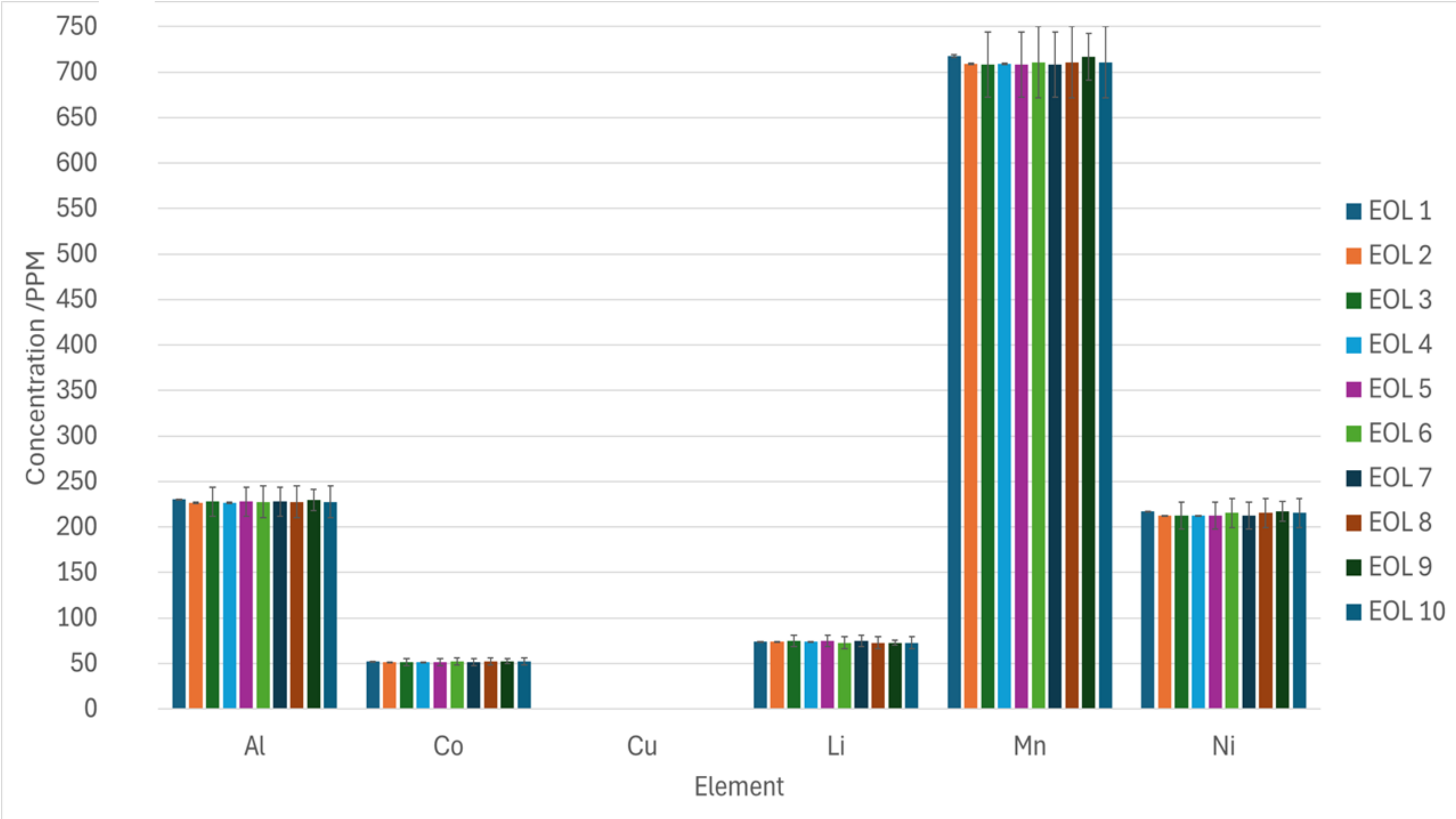


Figure 4.15: The variation in concentration of elements from 10 different samples from a single Nissan generation 1 cell cathode, with error bars from 3 repeats from each sample.

Table 4.7: The variation in concentration of elements from 10 different samples from a single Nissan generation 1 cell in PPM.

Sample	Al	Co	Cu	Li	Mn	Ni
EOL 1	230.48	52.41	0.01	73.72	717.77	216.97
EOL 2	226.63	51.43	0.01	73.67	709.03	212.24
EOL 3	228.03	51.46	0.01	74.93	708.47	212.78
EOL 4	226.63	51.43	0.01	73.67	709.03	212.24
EOL 5	228.03	51.46	0.01	74.93	708.47	212.78
EOL 6	227.45	52.17	0.01	72.68	710.77	215.35
EOL 7	228.03	51.46	0.01	74.93	708.47	212.78
EOL 8	227.45	52.17	0.01	72.68	710.77	215.35
EOL 9	229.49	52.60	0.01	72.70	716.93	217.28
EOL 10	227.45	52.17	0.01	72.68	710.77	215.35

Table 4.8 shows the mass of each element present in the sample in mg. Table 4.9 shows the percentage by mass of each element in the sample of cell cathode material.

Table 4.8: Mass of key elements determined by ICP-OES in the samples and the sample's corresponding weight.

Cell	Cathode Chemistry	Sample Mass (mg)	Al	Co	Cu	Fe	Li	Mn	Ni	P
Nissan Gen 1	NCA: LMO	546.30	57.62	13.10	0.00	0.00	18.43	179.44	54.24	0.00
Calb 130	LFP	454.90	66.39	0.00	2.39	105.61	12.89	0.16	0.07	60.44
LGM 50	NMC	454.30	30.56	22.52	0.02	0.00	21.12	11.03	192.67	0.04

Table 4.9: Percentage elemental makeup by mass of each sample. Determined from ICP-OES.

Cell	Cathode Chemistry	Sample Mass (mg)	Al	Co	Cu	Fe	Li	Mn	Ni	P
Nissan Gen 1	NCA: LMO	546.30	10.55	2.40	0.00	0.00	3.37	32.85	9.93	0.00
Calb 130	LFP	454.90	14.59	0.00	0.52	23.22	2.83	0.03	0.01	13.29
LGM 50	NMC	454.30	6.73	4.96	0.00	0.00	4.65	2.43	42.41	0.01

N.B. the elemental percentage does not add up to 100% as undigested organic material (i.e binder and conductive carbon) and oxygen are not detectable by ICP-OES.

The percentages in table 4.9 were worked out by calculating the mass of each reported element in mg from the concentration and then working that out as a percentage of the whole sample. Carbon would most likely come from conductive additive or binder, but the contribution of each cannot currently be determined and hence as can be seen in table 4.10 has to be labelled as "undigested material" rather than each component individually. Oxygen although important as part of the active material is not important to keep track of when not looking at direct recycling as the transition metals are going to undergo chemical processes to be remanufactured, and it is the recovery of the transition metals that determines the success of non-direct recycling method.

Figure 4.16 shows the SEM micrograph of the Nissan generation 1 cathode. It can be clearly seen that this consists of two sets of particles: the spherical NCA particles, and the angular LMO particles. It can be seen that the two are distinct and separate. Something that may be interesting to explore further would be to see if charge and discharge cycles, or in fact the over-discharge of the cell had an impact on the particle morphology. However, a fresh beginning-of-life sample that has not been over-discharged was not available for this comparison during this work.

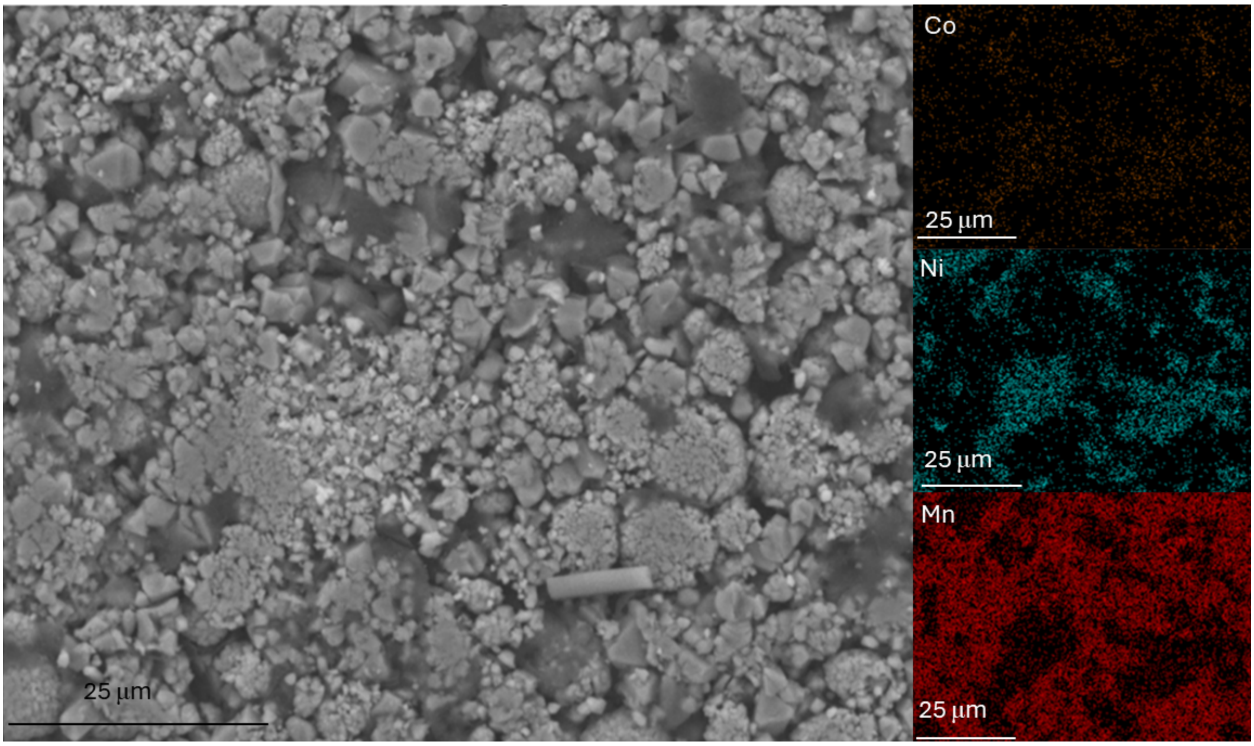


Figure 4.16: SEM micrograph of the Nissan generation 1 cathode showing two sets of distinct particles, NCA as highlighted by the overlapping Ni and Co EDX maps and LMO as highlighted by the Mn EDX map.

Figure 4.17 shows the SEM micrograph of the Calb 130Ah cathode, the rice-like particles of LFP can be seen clearly, with a homogeneous spread of Fe and P suggesting that the over-discharge of the cells has not damaged the particles at this level.

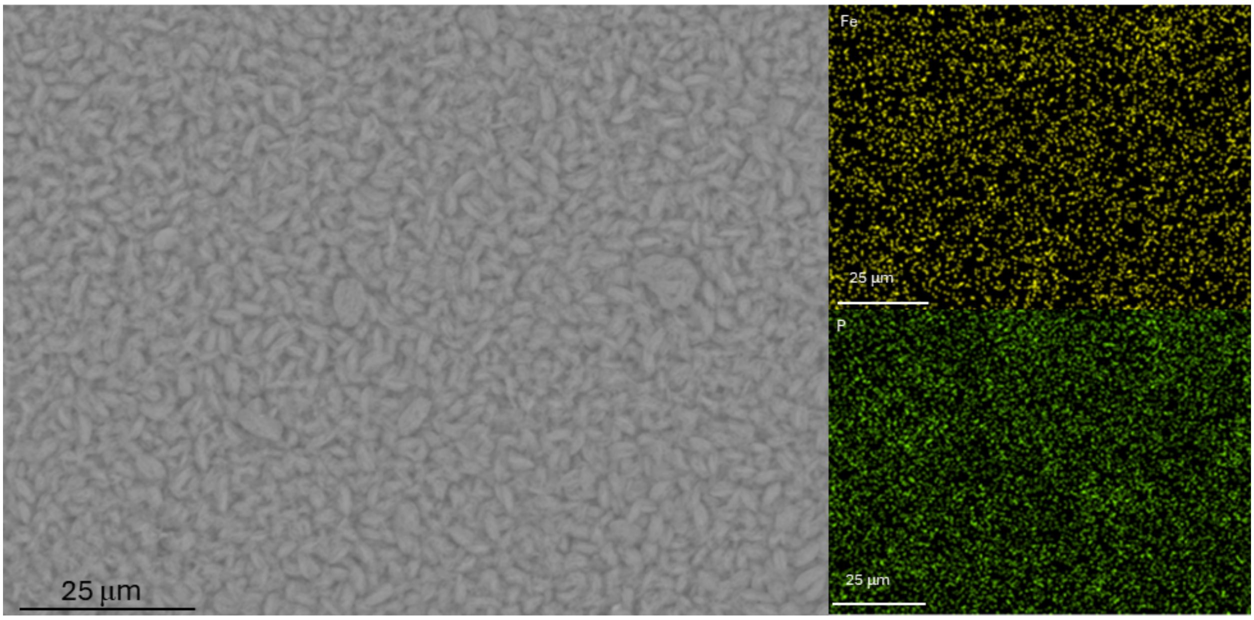


Figure 4.17: SEM micrograph showing the rice-like particles of LFP from the Calb 130 Ah cathode.

Table 4.10 shows how much of a sample of the cathode was not digested during processing for ICP-OES analysis. This gives a percentage by mass of organic components present in the cathode. These can be assumed to be split between conductive additives and binders. It is very clear that the LGM50 undigested material value is an outlier, as it would be expected to be in the 3-6% range. this is most likely down to error in the experiment during drying of the undigested part of the sample.

Table 4.10: mass and percentage make up of undigested material from cathode samples processed for ICP-OES analysis

Sample name	Mass of sample (g)	Undigested mass (g)	% Undigested sample
Nissan Gen 1 Cathode	0.29	0.02	5.94
LGM50 Cathde	0.21	0.0017	0.79
Calb 130Ah Cathode	0.22	0.01	3.28

Figure 4.18 shows the breakdown of individual components in the Nissan Gen 1 cell and their percentage by mass of the entire cell. "Binder+carbon+cathode" is the black mass component of the anode as the techniques used in this study are unable to distinguish between organic material. It can be seen that by mass the cathode makes up 50% of the cell, followed by the anode at 17.9%, and then the packaging, electrolyte and, separator make up the remainder. The results here agree with the results found by Marshal et al. [62], where the work found a 3.4:1 Mn:Ni ratio and these results found a 3.6:1 Mn:Ni ratio.

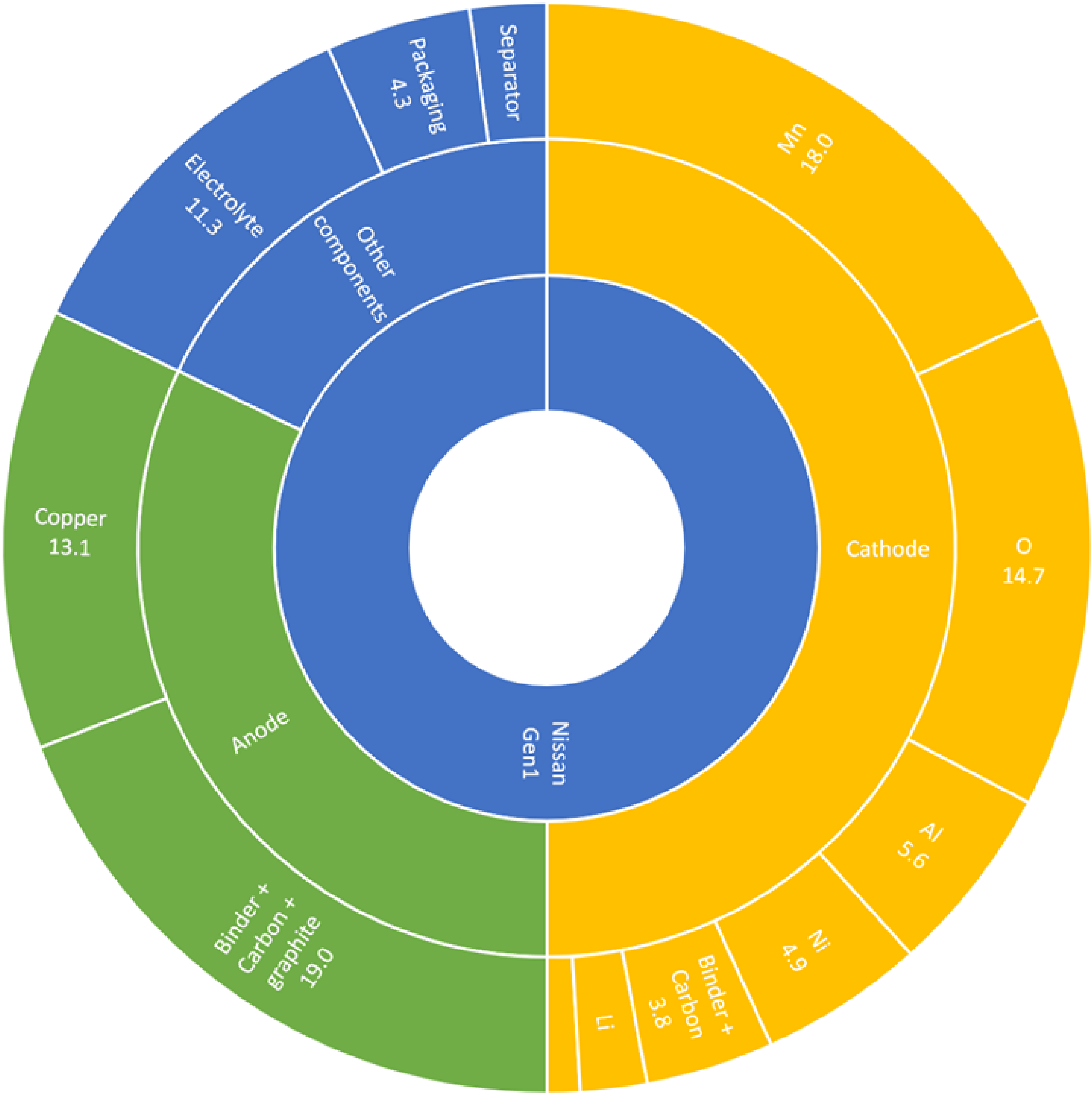


Figure 4.18: A breakdown of the individual components of the Nissan Gen 1 cell with their percentage make-up by mass of the entire cell. The cathode chemistry of this cell is LMO:NCA 75:25.

Figure 4.19 shows the breakdown of individual components in the LGM50 cell and their percentage by mass of the entire cell. "Binder+carbon+cathode" is the black mass component of the anode as the techniques used in this study are unable to distinguish between organic material. The cathode material makes up just under 41% of the cell, followed by the anode at 31%, and then the packaging, electrolyte and, separator make up the remainder. As discussed with respect to table 4.10 the reasoning behind the low undigested carbon/binder value is most likely down to error in the experiment during the drying of the sample.

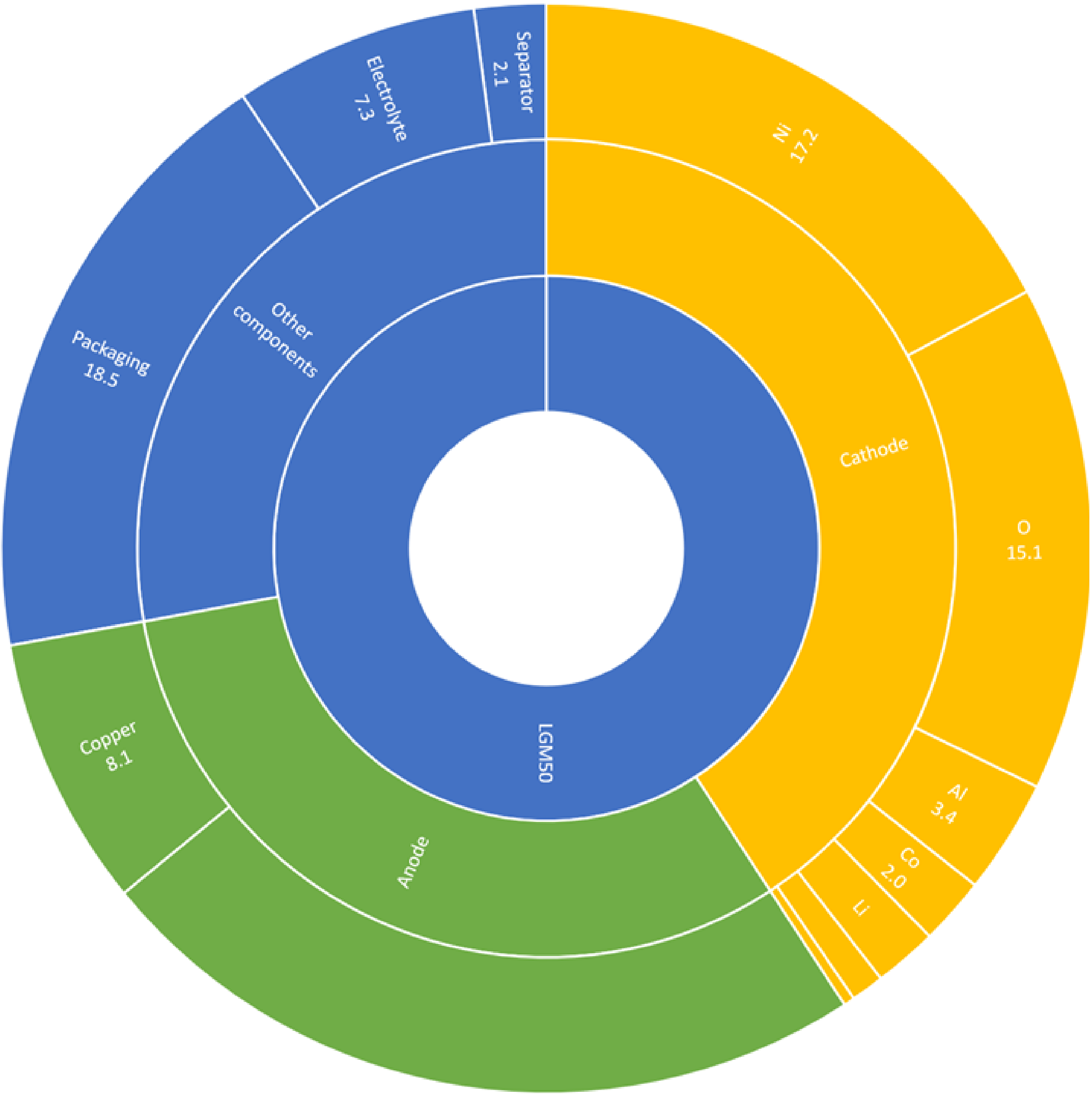


Figure 4.19: A breakdown of the individual components of the LGM50 cell with their percentage make-up by mass of the entire cell. The chemistry of this cell is NMC.

Figure 4.20 shows the breakdown of individual components in the Calb 130Ah cell and their percentage by mass of the entire cell. "Binder+carbon+cathode" is the black mass component of the anode as the techniques used in this study are unable to distinguish between organic material. The cathode material makes up just under 25% of the cell, followed by the anode at 24%, and then the packaging, electrolyte and, separator make up the remainder.

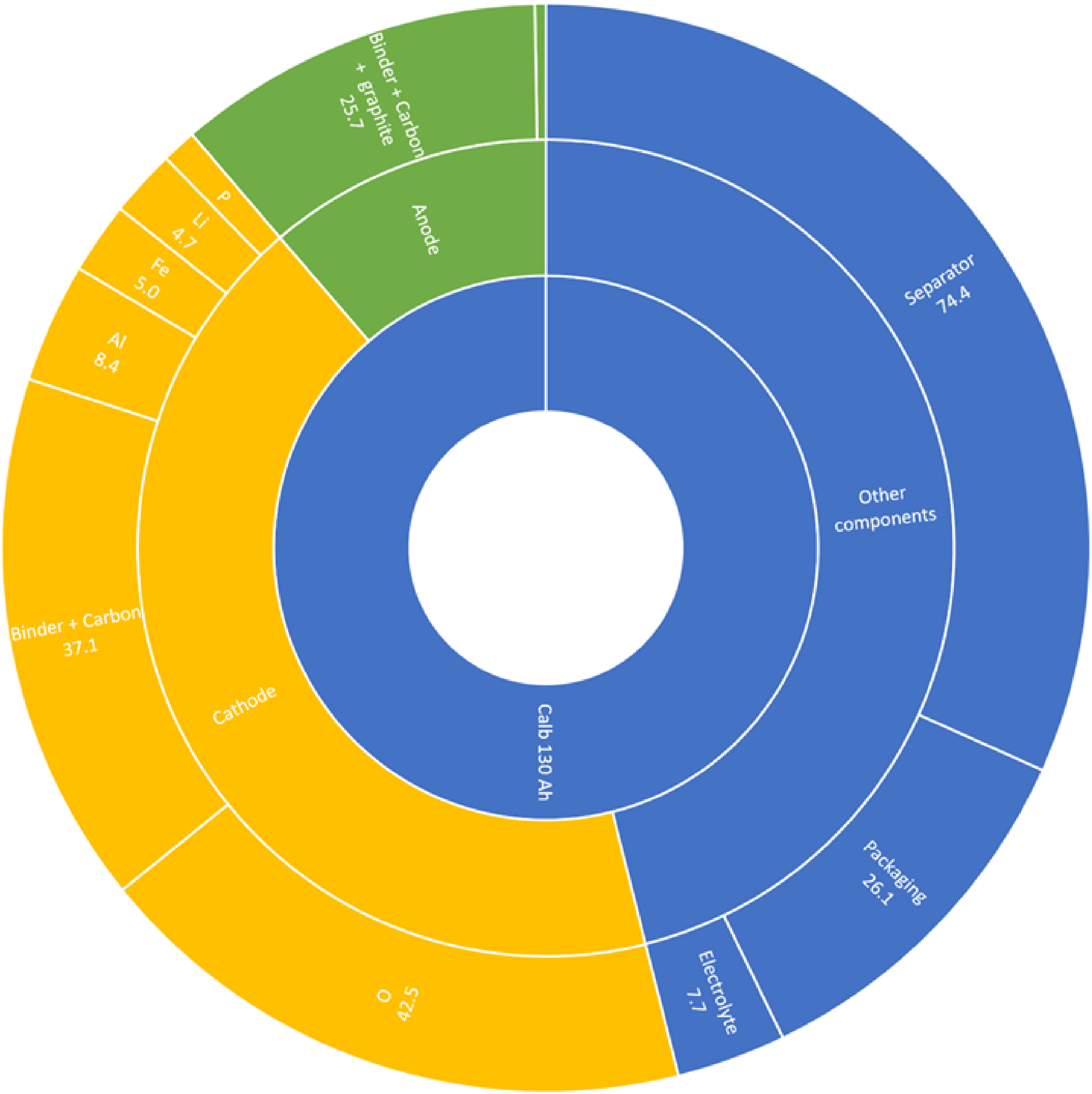


Figure 4.20: A breakdown of the individual components of the Calb 130 Ah cell with their percentage make-up by mass of the entire cell. The chemistry of this cell is LFP.

4.4 Discussion

4.4.1 Elemental and Materials Composition

As can be seen in Figure 4.15 the elements across the 10 samples that have been analysed have a very close clustering, with a range of no more than 2% between the highest and lowest measurement. This suggests that the digestion methodology is working efficiently and is digesting into solution all the present metals. If this was not the case, we would expect to see a wide fluctuation in the variability of elemental composition with no discernible pattern.

From the SEM images of the Nissan cathode (Figure 4.16), it can be seen that the cathode consists of two separate particles. An active material rich in Ni and Co and an active material rich in Mn. As the EDX for Mn does not overlap with the EDX Ni and Co this is unlikely to be an NMC chemistry and is, therefore, more likely to be a mix of NCA:LMO, this is confirmed also by the work done by Marshal *et al* [62]. With the SEM image of the Calb 130 Ah cathode (Figure 4.17), it can be seen there is a homogeneous dispersion of rice-like particles of LFP, Surprisingly, the EDX scan did not pick up any copper traces which are clearly present in the ICP results and are expected as the cells underwent an over-discharge to 0V. However, this could be due to the sensitivity of the EDX.

Comparing the composition of the cells, we can see that a benefit of the pouch cell is a much-reduced proportion by mass of packaging, with only 4.3% of the cell being packaging. However, the cells have to be arranged into modules that reinforce the cells before being assembled into the pack. Of the two rigid cells, LGM50 has a much lower weight component of packaging than the Calb cell. The Calb cell appears to have a lot more binder and conductive additive than the other two cells. From a recycling perspective, it is a detriment as the binder is difficult to remove and currently contains little value. From a valorisation perspective, NMC chemistries make the most sense to recycle as they contain high levels of Ni (17.2% by mass) and Co (2% by mass). The LGM50 cell has a water-based binder on the anode. This is a step forward in the recycling potential as no expensive or harmful solvents are needed. However, the cathode is still PVDF, which is true for both electrodes in the other two cells. Z-fold (like in the Calb cell), or the jelly roll (like in the LGM50 cell) make for far easier disassembly of the jelly roll or stack, than the stacked construction of the Nissan cell. If an appropriate solvent is chosen to separate black mass from the current collector, the retention of the current collector in its metallic form is very feasible for all cell designs and is further investigated in Chapter 6.

4.4.2 Safety

Before commencing any disassembly, it is important to ensure that good lab protocols, including the use of appropriate PPE (such as lab coats, goggles, and gloves), adhering to risk assessments, COSHH assessments, and training are conducted and understood. The work should also be performed using appropriate engineering controls such as under extraction as the electrolyte contains volatile organic components and toxic additives that should not be inhaled. On a lab scale, this could be a fume-hood or a glovebox if electrochemical characteristics need to be preserved. However, on an industrial scale if the cells are being manually disassembled appropriate PPE may have to include a self-contained breathing apparatus similar to hazmat suits. Disassembling a lithium-ion cell is an inherently risky process. In this study, many things have been considered to account for safety and attempt to prevent unexpected events. Firstly all cells must be, as a minimum, discharged to a voltage where they are at a 0% state of charge. This is between 3.0 V and 2.5 V [122]. Another novel methodology that has been used for the opening of cylindrical cells has been to use laser ablation [137]; however, a downside of this method is that due to the high levels of heat generated, it may not be possible to use cells where the separator is sealed within the seal as this would not open the cell due to the melting of the separator. This does not entirely remove the risk of a thermal event as 0% state of charge is not the same as 0 V [122]; however, it should reduce the severity of an event. 0 V is not used as, although this is safer, it contaminates the materials with copper from the anode current collector as the voltage at the negative electrode increases, and the copper oxidises and subsequently dissolves into the electrolyte, re-precipitating when the cell is recharged, or the external circuit is removed [138]. Ideally, x-ray tomography would be carried out before disassembly to ensure that the internal layout of the cell is understood and the gaps in the assembly where it is safe to cut are identified. X-ray scanning of a cell is both time-consuming and space-restricted so can probably only be done during the cutting pattern development stage of standardised cells. If this is not possible, the supplier and, in future, the battery passport should, at a minimum, describe the chemistry of the cell (both anode and cathode), which should be confirmed with a charge/discharge cycle that would also put the cell at 0% charge, in order to minimise the scale of any thermal event. Also, the disassembly should be conducted in such an area that can be easily and quickly evacuated, and secured to contain the event, and the thermal properties of the cell should be continuously monitored.

Ceramic blades should be used where possible when cutting into any cells. This mitigates the risk of a short circuit if the stack or jelly roll is nicked during the incision process by removing a conductive path (the blade) via which a short circuit could occur. Other methods such as carbide saws [139], or band saws (as demonstrated by KYBURZ AG [133]) can also be used. As discussed in the introduction, cells contain several volatile organic compounds as part of the electrolyte, such as dimethyl carbonate, ethyl methyl carbonate, ethylene carbonate, and other additives [22, 140, 141] therefore all disassembly work must be done under adequate extraction, not only does this prevent the inhalation of compounds hazardous to health but also prevents the build-up of a potentially flammable atmosphere. Also, when cutting, it is essential to cut into voids and avoid hitting the electrode stack/jelly roll.

Finally, concerning the storage of samples, in later chapters, "aged" material will be used, defined as material that is exposed to the atmosphere for a minimum of 4 weeks. These samples are all washed in isopropyl alcohol for a minimum of 8 hours before being stored or undergoing any further characterisation. The reasoning behind this is that the electrolyte that should have fully penetrated all pores in the electrodes contains fluorine-containing components (e.g $LiPF_6$) which may decompose to HF [142] the washing process should remove the salt from the electrode, removing this hazard. After the washing, the components are heated and vacuum-dried to remove any remaining isopropyl alcohol (IPA) and any reaming electrolyte solvents that may be within the pores of the electrodes. If the samples must be stored for extended periods, they should be stored in vacuum-sealed pouches. This does not eliminate degradation of the material; however, it does retard it and extends the shelf-life of the material. Hence, when designing recycling operations, it should be considered that the opening and sorting facility should be as close as possible to any other process if the preservation of material structure is important so that shipping does not add any considerable time and does not allow the material to degrade.

4.4.3 Design for Disassembly, Recycling, and Future Trends

The methods and concepts of safe quantifiable disassembly presented in this chapter apply not only to the cell geometries described herein but also to future cells. To that effect, this has been applied to two other cells: a modern commercial prismatic cell from a vehicle less than 10 years old and a super-capacitor.

A few things can be observed about the modern cell compared to the Calb cell. The preference for welding rather than mechanical fastening as the attachment method between tab and tag (Figure 4.21a), negatively impacts the ability to recycle cells as the detachment of external packaging from electrode stacks wastes current collector material. External packaging material has shifted from plastic to metal from a recycling viewpoint this is an improvement as metals are easier to deal with than plastics as metal identification is both easier and they are more commonly recycled, whereas some plastics are not commonly recycled. However, from a disassembly viewpoint metals are more resistant to cutting so require more effort to get to the electrode stacks. Also, there has been a marked decrease in the so-called "dead space" within the cell in which to cut, meaning that tomography may be required for proper safe disassembly of the first cell (Figure 4.21b). Another observation from the more modern cell vs the older prismatic cell is that the safety vent in the modern cell is integrated and is a place for the cell to rupture, whereas, in the Calb cell, it is more like a cap with a spring valve in it that can be removed. This means that the Calb cell is easier to drain off electrolytes than the more modern cell.

From a design for disassembly perspective, this study has focused on the cell level rather than the pack or module level. Although much can be said for further improvement in the disassembly of packs and modules, it is out of the scope of this work. Regarding individual cells, disassembly rather than shredding allows for the recovery of components with minimal damage. This allows for higher purity in the recovered material than shredding and minimises the sorting required, as this can be done in one step during disassembly. This is because, in disassembly (manual or automated), the components are not broken down and mixed. Another benefit of disassembly is that it is possible to remove electrolytes by washing the separated components without cross-contamination of materials, which negatively impacts the quality of the recovered material [143]. In turn, not only does this potentially shorten the process but also leads to better valorisation of valuable materials such as Ni, Co, and Li [3, 144, 145]. However, a downside to disassembly, at least on an industrial level, is that this is a considerably slower process as each cell would need to be addressed individually, whereas it is possible to shred entire packs from a vehicle.

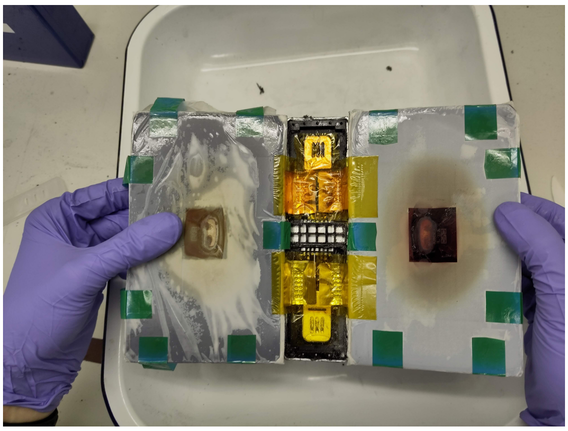
So what makes for a good cell from the disassembly and material recovery aspect? Starting from the casing, this needs to be a material that is easy to penetrate deliberately, and this can be done in several ways: including a deliberate failure point that becomes vulnerable under specific conditions (for example, exposure to a unique wavelength of radiation such as UV that after construction the internals of a pack would not be exposed to), having specific dead zones into which a blade or saw can cut into without damaging the electrode stack (such as the large gap seen at the top of the Calb cell), or be made of a material that is tough enough for the cell but is easy to puncture like the pouch material seen in the Nissan pouch cells. Also, there must be an easy way to discharge all the cells to a 0% state of charge, be that at the pack, module, or cell level.

Once the electrode stack is reached, how it is constructed is also important. A roll such as seen in the LGM50 cell is easy to disassemble as it can be simply unwound and the anode and cathode are separated as single long sheets. However, due to the casing design, the jelly roll is difficult to extract and requires the majority of the can to be removed. A good alternative, especially if using a prismatic or pouch casing, is the z-fold operator as used in the Calb cell, allowing for a machine to grab and wind up the separator with the anode and cathode being ejected into separate bins. A stacked stack, such as seen in the Nissan pouch cell, is the least favourable as it requires each layer to be dealt with separately.

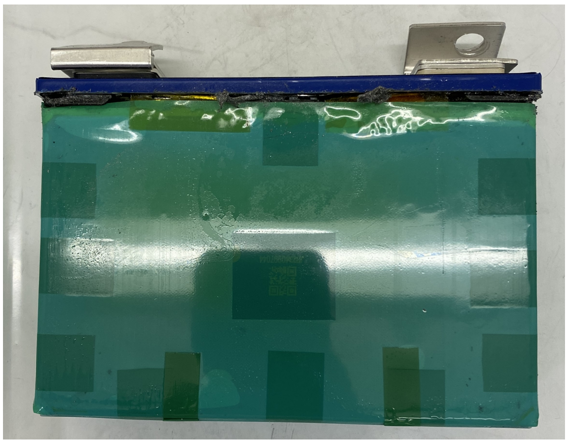
Once the components have been separated, the black mass must be recovered. Firstly, it is best to keep the anode and cathode separate to prevent cross-contamination, as discussed previously. For black mass recovery and valorisation, it is important to retrieve as pure an output as possible, which means removing any binders and conductive additives. It would be beneficial if the binder systems were soluble in a benign solvent such as water. This move has been seen on the anode side but is still a hurdle on the cathode side, which mainly uses

PVDF, however, research is ongoing [146–148]. Also, for separation purposes, the conductive additive should have at least one physical property that differs considerably from the active material, such as hydrophobicity or reaction to external electromagnetic fields. The recovery of electrolyte components, especially those containing lithium, would benefit a zero-waste process; however, this was not investigated in this work.

Once the active material, binder, current collector and any additives have been separated they can then be reused in the manufacturing process. For this to be true for direct recycling (as defined in the introduction) the active material must be chemically, electrochemically, and morphologically indistinguishable from virgin material, this includes not having a reduced lithium content for the material to be suitable for re-incorporation without re-lithiation. This indistinguishability is not so important for the current collectors as long as they remain in the metallic form as these can be recycled with their respective bulk component pyro-metallurgical.



(a) Attachment between tab and tag using ultrasonic welds under Kapton tape.



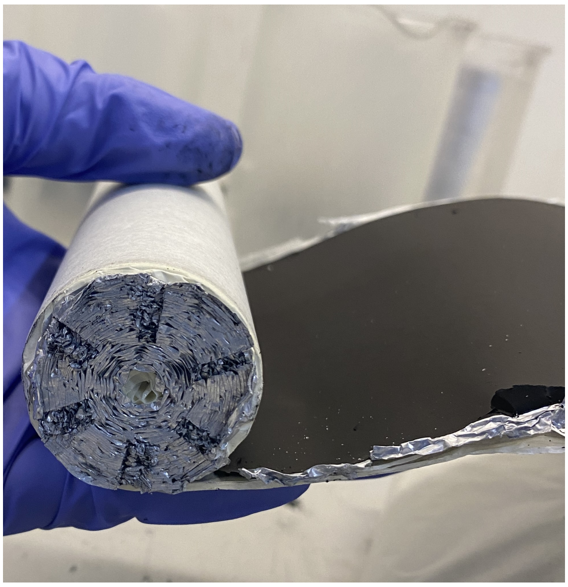
(b) Electrode stack from a modern commercial cell removed from metal casing. As can be seen, there is minimal dead space.

Figure 4.21: Images of a modern commercial cell showing: a) the sonic welds used to attach electrodes to terminals, b) the tight packaging of the stack within the case..

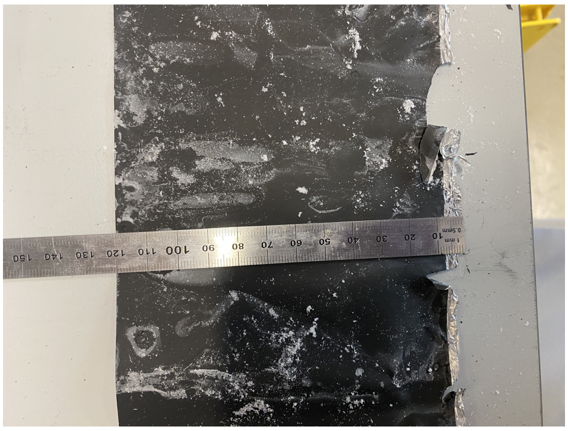
As an interesting test case, a proof of concept that the disassembly methodology could be applied to other electrochemical storage devices, and training exercises, a cylindrical super-capacitor (Figure 4.22a) was also disassembled using the concepts developed here. It was found that the capacitor had a tab-less design. This is currently being investigated commercially by cell manufacturers such as Tesla for lithium-ion cells. This reduces the dead space in the cell compared to a metal-encased prismatic. However, from a recycling and disassembly perspective the opening procedure becomes somewhat more difficult, as instead of having one tab to tag weld, there are multiple (dark areas in an asterisk pattern Figure 4.22b and 4.22c) also the unrolling of the jelly role becomes more difficult potentially leading to reduced yields as the current collector is prone to ripping as seen in Figure 4.22d.

(a) Exterior of the super-capacitor with scale.

(b) Roll facing face of the super-capacitor end cap with weld points appearing as darkened areas.



(c) End on view of the super-capacitor jelly roll with tab-less design.



(d) Unrolled super-capacitor electrode showing tears along tab-less design edge.

Figure 4.22: Images of aspects of a tab-less design super-capacitor: a) Exterior image showing the size, b) end cap showing the connection method of a star shape set for welds, c) Image of the jelly roll end on showing tab-less current collector d) width of the electrode showing several tears that occurred during the unrolling process.

4.5 Conclusion

To conclude, this study has developed a suite of analysis techniques and principles to safely and quantifiably disassemble a cell, and then analyse the components down to an elemental level. The process consists of entering the case via incisions into empty spaces in the cell, disassembly, weight analysis, ICP-OES, SEM-EDX, and XRD analysis. This process has been applied to three of the most common automotive cell designs; pouch, cylindrical, and prismatic. This has also been applied to a much more modern prismatic cell with a successful disassembly, and to a super-capacitor showing that at least the disassembly methods have a wide range of applicability to the disassembly of energy storage devices.

Having disassembled three of the most common large-scale commercial cells and discussed the benefits and drawbacks of each cell construction, the following suggestions would make for the most recyclable cell:

1. the outer case: the case should be either a pouch or prismatic cell with a predefined and triggerable failure point.
2. the electrode stack: should be either a roll or a z-fold stack
3. the electrode coating: should be bound with water-based (or benign solvent) binders, and contain as few additives as possible that have at least one distinct physical property for separation.

It does not escape the author that the recommendations made here are somewhat at odds with making the best-performing, cheapest, easiest-to-manufacture, most profitable cells. Things like a failure point and having "dead space" in a cell potentially reduce lifetime and/or energy density, more complex stacking arrangements increase manufacture time and complexity, and developing novel electrodes and binder systems requires capital investment cutting into profit margins. However, there is no point in having a cell that stores hundreds of kWh of energy, can be manufactured in seconds, and costs pennies if the other cost of this is environmental ruin. Industry has always and will continue to innovate around legislation, and it is the role of governments to ensure that legislation is there to ensure that the green credentials of electrochemical storage devices are improved and benefit society as a whole and not simply corporate bottom lines.

Now that the composition of the cells that are available for recycling is understood and quantified, the development of the recycling process can begin as it will now be possible to assess the efficacy of these processes, track any waste that may be occurring, and understand where value is being retained. In the following chapters two processes will be investigated, each looking at enabling greater value retention within the recycling process.

Chapter Five

Acid Assisted Separation of Cathodic Black Mass from Current Collector

This work has been published as a paper in the journal Metals: Zorin, A., Song, T., Gastol, D. & Kendrick, E. Acid-Assisted Separation of Cathodic Material from Spent Electric Vehicle Batteries for Recycling. en. Metals 13, 1276. issn: 2075-4701. https://www.mdpi.com/2075-4701/13/7/1276 (2024) (July 2023). The author's contributions are as follows: A. Zorin - lead author ICP, electrochemistry, SEM, acid delamination data acquisition, and data analysis and visualisation, E. Kendrick - Supervision, Reviewing, and Editing. D. Gastol - Electrochemical methods standardisation and cell construction, T. Song - XRD acquisition and analysis.

5.1 Introduction

The recycling of lithium-ion batteries presents challenges due to the complex composition of waste streams generated by current processes. Achieving higher purity levels, particularly in the reclamation of aluminium metal and transition metal black mass, is essential for improved valorisation. This study proposes a high-efficiency, low-energy, and environmentally friendly method using organic acids to separate the cathodic black mass from the aluminium current collector. In this work, several mild acids (oxalic acid, citric acid, malic acid, lactic acid, acetic acid, succinic acid, and pimelic acid) with pKa >1 were chosen as the potential acids for direct recycling. For this process, a low molar concentration (0.5 M) of acid was used to maximize the metallic aluminium recovery. Mechanical agitation was used to expedite the separation process by ensuring that the solution was able to contact all of the cathode strips and break off black mass from the bulk once it had separated from the aluminium current collector. As a result, sufficient separation was achieved. This organic acid separation process has great potential as a concentration step for cathode electrodes, providing feedstock for subsequent direct or short-loop recycling. The acids selected in this study all show >86% black mass recovery with acetic acid showing 100% black mass recovery of black mass from the current collector. The recovered materials were subjected to X-ray diffraction, electron microscopy, and elemental analysis techniques. this work shows that oxalic-acid-treated material exhibited two distinct active material components with a minimal change in mass ratio compared to the untreated material. This work shows by elemental analysis of the leachates that the majority of critical materials were retained in the black mass and limited aluminium was leached during the process, with almost 100% of Al recovery achieved. This methodology enables the production of high-purity concentrated aluminium (which although low value as seen in Figure 2.2, is incredibly energy intensive to recover when not in its metallic state) and critical metal feedstocks (Mn, Co, Ni, and Li) for further hydro-metallurgical processes, upcycling of the cathode material, and direct recycling. The proposed approach offers significant potential for enhancing valorization in lithium-ion battery recycling, facilitating efficient separation and optimal recovery of valuable metals.

5.2 Methodology

The electrodes used in this study are harvested from manually disassembled 1st generation Nissan Leaf cells, the active material is $LiMn_2O_4/LiNi_xCo_yAl_zO_2$ (LMO/NCA lithium manganese oxide/nickel cobalt aluminium oxide) $x+y+z=1$ with a ratio of 3:1 with PVDF as binder [62]. The weak organic acids are used in this work to maximize the metallic aluminium recovery, limit the level of aluminium dissolved into the leachate, and preserve the transition metal and lithium content in the cathode materials. This organic acid separation process has great potential as a concentration step for cathode electrodes, providing feedstock for subsequent direct or short-loop recycling.

To separate the active material containing black mass from the current collector, the cathode electrodes were first obtained by manual disassembly from a 1st generation Nissan Leaf cells, the active material is $LiMn_2O_4/LiNi_xCo_yAl_zO_2$ (LMO/NCA) $x+y+z=1$ with a ratio of 3:1 with PVDF (Polyvinylidene Fluoride) as binder. The electrodes were shredded into uniform rectangle pieces of approximately 2 cm x 5 cm. The mass of each strip was recorded to ensure uniformity for each test. An acid solution was preheated to 50 ℃ on a hotplate (Heidolph Instruments, Germany) as was the mixer vessel. A 0.5 M solution of the acids was prepared. The acid solution was placed into the mixer vessel followed by the cathode pieces in a 10:1 liquid-to-solid ratio. The overhead mixer was set up to stir the sealed vessel at 300 RPM for 5 minutes. After the mixing time, the acid solution was immediately filtered, and the collected filtered acid solution was used to wash the mixer vessel to ensure all liberated material was removed. The material was then dried at 60 ℃ overnight in an oven.

black mass recovery was used to quantify the success of separation, which was defined as below:

$$E_a = \frac{mass\ of\ separated\ component}{total\ theoretical\ mass\ of\ cathode\ electrode\ coating} \times 100\% \qquad (5.1)$$

where E_a is black mass recovery in %; the theoretical mass of cathode electrode coating is worked out to be 86% by mass from delaminating work in Chapter 4.

A baseline for performance and comparison of an end-of-life (EoL) was also taken.

Material characterisation and electrochemical testing are described in detail in Chapter 3

5.3 Results

The performance of the selected acids (oxalic acid, citric acid, malic acid, lactic acid, acetic acid, succinic acid, and pimelic acid) was assessed for separation efficiency and black mass elemental content.

To compare the effectiveness of the process, the amount of transition metals leached and the lithium content were compared in the recovered black mass and in the leaching solution (PPM results can be seen in table 5.2, the slight variation of aluminium in the black mass likely comes from some contamination of the black by aluminium leached from the current collector and does not suggest greater than 100% peeling). The % content of the black mass is illustrated in Figure 5.1a. For the theoretical cathode material, it was assumed that the material is 75:25 $LiMn_2O_4$ and NCA, respectively based on the work presented in the previous chapter and as also found by Marshal et al [62]. From this, the elemental mass was calculated. This was compared to the elemental content in the untreated and acid-delaminated materials. It is clear that all the metals present in the cathode strip are being leached. This is likely due to the fact that the organic acids act both as leaching

Table 5.1: Summary of the mass of black mass and Al current collector collected after separation.

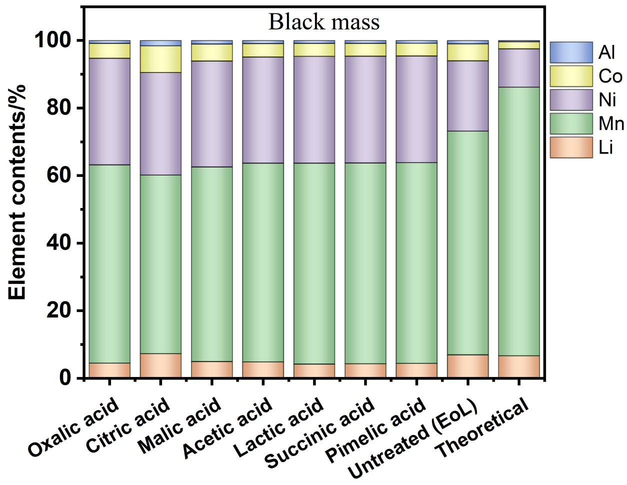
Acid	**Mass of Electrode Sample (g)**	**Mass of Separated Black Mass (g)**	**Al Mass (g)**
Oxalic acid	7.513	6.897	1.157
Citric acid	7.485	3.552	1.476
Malic acid	7.510	5.000	1.614
Acetic acid	7.517	5.563	1.494
Lactic acid	7.487	4.724	1.416
Succinic acid	7.488	5.085	1.742
Pimelic acid	7.519	5.000	1.786

acids and reducing agents, reducing the metals from their insoluble to soluble states [42–45]. Manganese is perhaps preferentially leached as it forms the most stable complex as suggested by the Irving-Williams series [149]. All the acids used during this study leach lithium to some extent, which is expected.

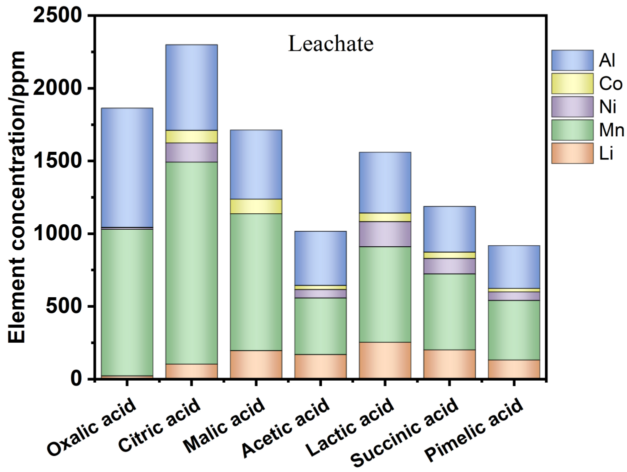
The XRD patterns of the recovered powders from the acid treatments show similar patterns compared to pristine untreated material (Figure 5.2a). However, minor changes in the main characteristic peaks belonging to LMO and NCA can be observed in the enlarged patterns. The changes in peak positions and intensities can be explained by the loss of metals due to acid leaching. Previous reports have mentioned that acids are selective for the leaching of metals, leading to the difference in the final composition and crystal phase [150]. To confirm this, Rietveld refinement was conducted using the GSAS software, and the results are shown in Figure 5.2b–h and Table 5.3. The pristine cathode is composed of LMO and

Table 5.2: Elemental the makeup of black mass in PPM.

Element	Oxalic	Citric	Malic	Acetic	Lactic	Succinic	Pimelic
Al	10.5	12.8	11.9	12.0	11.7	11.8	11.3
Co	56.2	65.1	59.7	54.9	55.5	55.1	54.5
Mn	747.2	436.2	679.7	806.6	858.7	862.1	857.0
Ni	401.7	250.6	369.7	430.8	457.1	458.6	455.7
Li	57.5	60.4	58.7	66.8	61.3	62.6	64.4



(a) The % mass contribution in the black mass



(b) The level of leaching from the coated current collector

Figure 5.1: ICP-OES analysis of metal content with a) the % mass contribution in the black mass and b) the level of leaching from the coated current collector when delaminated.

NCA in a 3:1 ratio. Table 5.3 shows that the phase fraction of powder treated with oxalic acid is the closest to the designed ratio of 3:1, which means that the fewest metals were leached during the separation process. This was also verified by the ICP data discussed above. Powders treated with citric acid, malic acid, and acetic acid maintain their original structure. Significant changes in the crystal structure were observed after treatment with lactic acid, succinic acid, and pimelic acid.

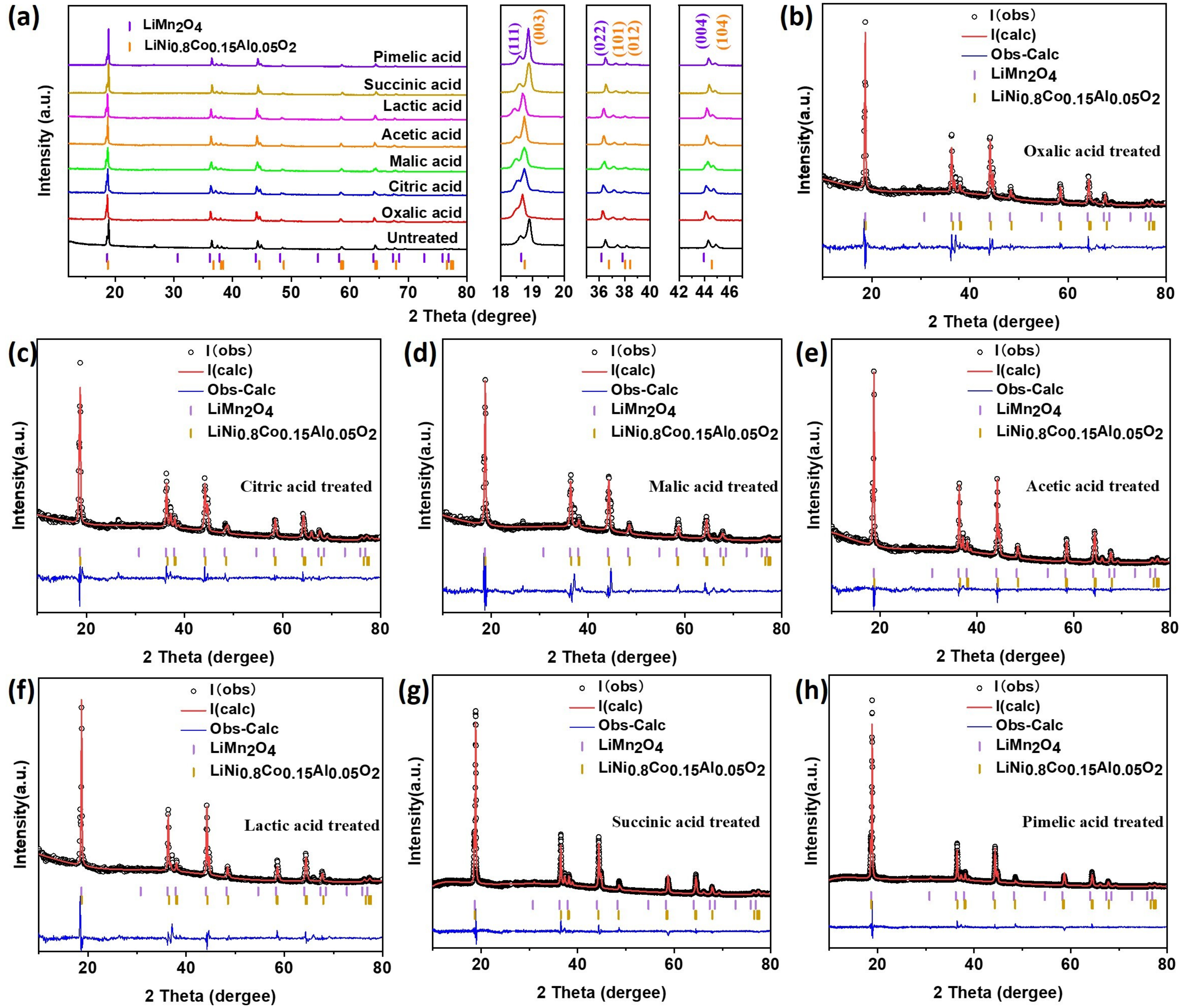


Figure 5.2: XRD patterns of a) the recovered material and untreated material as a comparison, b) oxalic acid treated pattern, c) citric acid treated pattern, d) malic acid treated pattern, e) acetic acid treated pattern, f) lactic acid treated pattern, g) succinic acid treated pattern, h) pimelic acid treated pattern.

Table 5.3: Summary of material weight fractions in acid processed materials by XRD.

Acid	**LMO Wt.Frac.**	**NCA Wt.Frac.**	**Ratio Wt. Frac. 1/2**	**Rwp %**	**Rp %**	**Chi^2**
Oxalic acid	0.75264	0.24736	3.04	5.02	3.21	10.15
Citric acid	0.75918	0.24082	3.15	5.90	4.08	4.577
Malic acid	0.76608	0.23392	3.27	6.21	3.48	15.63
Acetic acid	0.76843	0.23157	3.32	3.64	2.54	4.777
Lactic acid	0.81942	0.18058	4.54	5.72	3.27	11.82
Succinic acid	0.80778	0.19222	4.20	4.12	2.91	1.07
Pimelic acid	0.82649	0.17351	4.76	4.27	2.97	1.23

Figure 5.3 shows the SEM images of the processed powders and EDS maps of Mn and Ni. From the figures and maps, it can be seen that there are two distinct particles. The first is a manganese-rich phase which we can assume to be LMO; this is smaller and more angular than the second particle type, which is larger and spherical. This second particle is nickel, rich with low levels of aluminium and cobalt, as shown in the EDS maps.

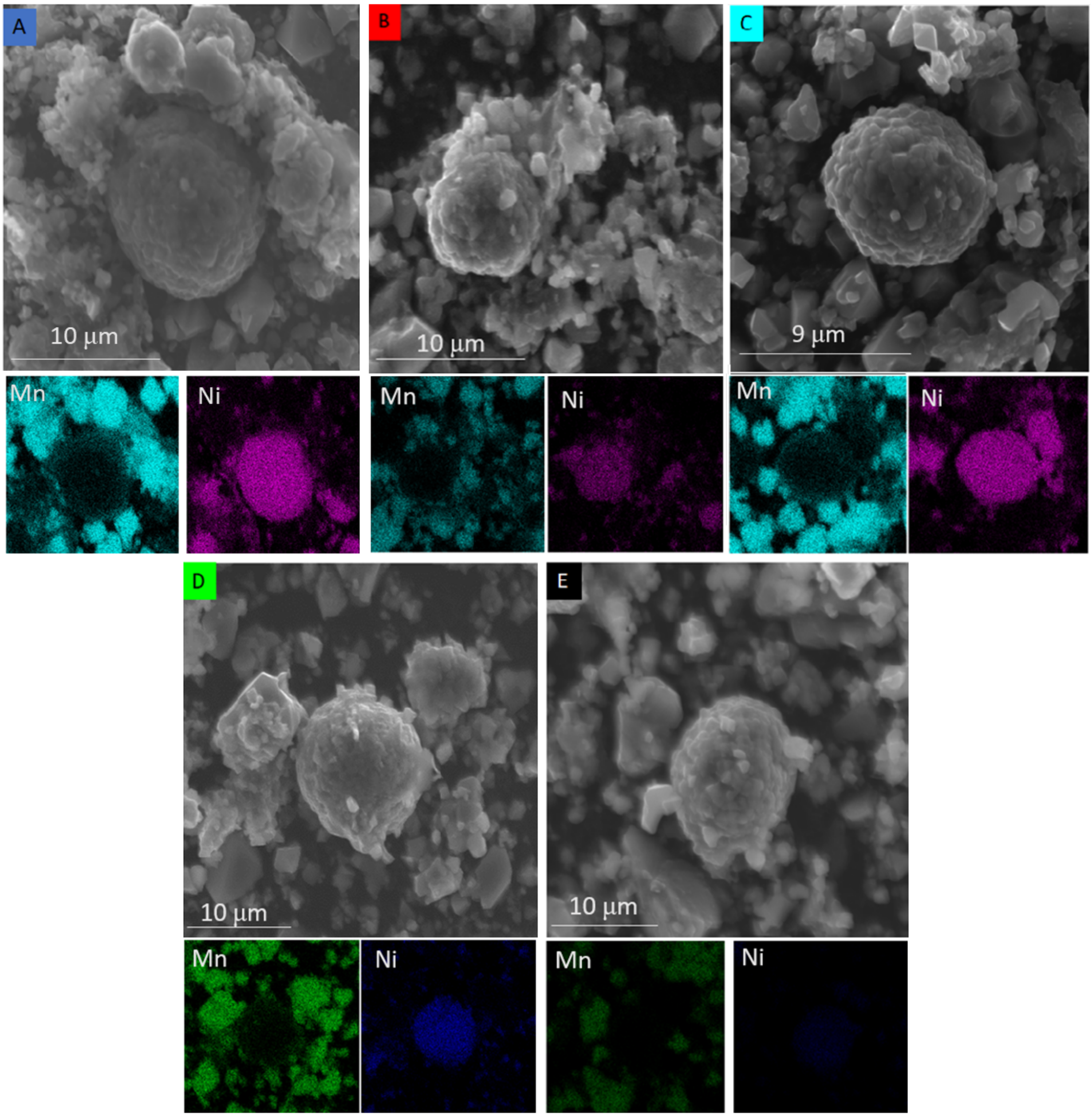


Figure 5.3: SEM micrographs of treated black mass powders with selected elements: a)acetic, b) citric, c) lactic d) malic, and e) oxalic acid. Manganese in teal and green. Nickel in pink and blue.

The recovered black mass was not suitable for direct recycling without further treatment because its capacity did not reach or exceed that of the untreated end-of-life material, as can be seen in Table 5.4. However, the samples were tested electrochemically to determine the retained capacity in the reclaimed cathode. The cathodic black mass was made into an electrode, as described in the electrochemical testing section of methods after drying at 120 ℃ for 8 h. Table 5.4 shows the first-cycle capacities of the processed materials, the peeling efficiencies of the chosen acids, and an untreated baseline material (EoL Unprocessed). It was observed that all the acid-processed materials had some capacity loss when compared to the unprocessed material (the fact that the coatings were handmade rather than mass-manufactured could also contribute to this). This is likely due to transition metal and lithium leaching from the active material. However, the capacity, as measured electrochemically, compares well to the lithium content measured with ICP-OES measurements. Acetic acid, which leached the least Li when compared to the EoL untreated sample, showed the highest capacity retention when compared to the unprocessed material of 84.51 mAh/g (78.5% vs EoL unprocessed). Although the general trend does appear to be that Li retention is key to capacity retention after processing there is an outlier to this trend; which is oxalic acid as it only retained 57.49 PPM of Li but had a first-cycle capacity of 69.71 mAh/g which is the second highest of the processed materials. The smallest hysteresis between the charge and discharge parts of the cycle has also been shown by the acetic acid processed material as can be seen in Figure 5.4. All the other liberated cathode materials exhibited specific capacities between 40 and 60 mAh/g, with considerably larger hysteresis between charge and discharge due to lower lithium content in the materials. Acetic, lactic, and succinic acids had the highest black mass recovery, with 100%, and from the ICP-OES analysis of the leachate solution, the lowest levels of aluminium present (acetic: 372 ppm, lactic: 417 ppm, succinic: 313 ppm), suggesting that these acids do the least damage to the current collector upon delamination.

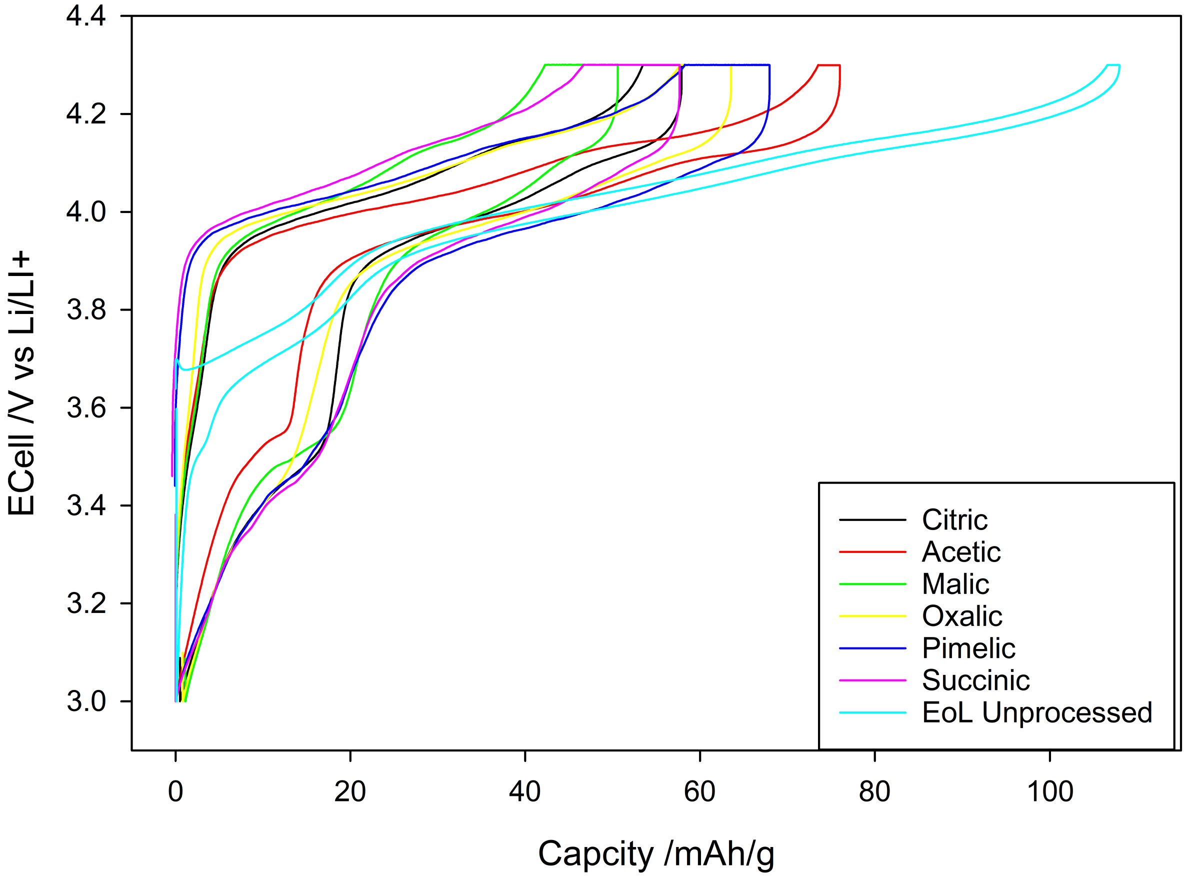


Figure 5.4: First cycle capacities of acid treated material and EoL unprocessed material, showing a decrease in capacity of acid treated material vs EoL material.

Table 5.4: First-cycle capacity and black mass recover.

Processing Acid	First-Cycle Capacity (mAh/g)	black mass recover (%)	Li^+ Content (ppm)
Citric acid	57.32	99.5	60.44
Acetic Acid	84.51	100.0	66.76
Malic Acid	54.23	86.4	58.66
Oxalic Acid	69.16	96.53	57.49
Pimelic Acid	67.26	100	64.41
Succinic Acid	60.15	100	62.57
Lactic Acid	66.04	96.50	61.26
EoL Unprocessed	107.62	N/A	71.71

The processing of electrodes with organic acids for delamination of cathode active materials from the aluminium current collector offers an effective method for separating black mass from the current collector with separation efficiencies of no less than 86%. Acids such as acetic and pimelic acid can achieve complete separation, but leach transition metals into the acidic solution. This process can serve as a crucial pre-concentration step in recycling efforts. While the choice in acid does have an impact on separation efficiency, all tested acids in this study demonstrated peeling efficiencies greater than 85%, as shown in Table 5.4. Additionally, the choice of acid influences the quality of the recovered black mass, as certain acids preferentially leach transition metals. For instance, citric acid retains the least amount of manganese (436.2 ppm)and nickel (250.6 ppm) in the black mass.

Comparing organic acid separation it can be seen that this performs similarly to mineral acid separation (<84% black mass recovery [53]), somewhat worse than solvent separation (<99% black mass recovery [54]). However, when comparing organic acid separation to thermal treatment and cryogenic grinding it compares favourably as these achieved lower black mass recovery (<50% and <73%respectively [95, 100]).

5.4 Discussion

It has been shown that organic acids can be effective in separating cathode black mass from aluminium foil [62] by pacifying the aluminium film from corrosion, creating a superphobic organic layer [151–153] on the surface which then repels the water, creating bonds between the current collector and binder. They also selectively leach lithium, transition metals, and aluminium, which weakens the chemical bond between the aluminium foil and the binder in the cathode black mass [154–159]. Equations (5.2)–(5.4) represent the typical dissolution reactions by taking $LiNi_{1/3}Mn_{1/3}Co_{1/3}O_2$ and acetic acid as an

$$CH_3COOH(aq) \rightarrow CH_3COO^-(aq) + H^+(aq)\ pKa = 4.75 \tag{5.2}$$

$$2Al(s) + 6CH_3COO^-(aq) + 6H^+(aq) \rightarrow 2Al(CH3COO)_3(aq) + 3H_2(g) \tag{5.3}$$

$$\begin{gathered} 3LiNi_{1/3}Mn_{1/3}Co_{1/3}O_2(s) + 9CH_3COO^-(aq) + 9H^+(aq) \rightarrow \\ Ni(CH_3COO)_2(aq) + Co(CH_3COO)_2(aq) + Mn(CH_3COO)_2(aq) + \\ 3Li(CH3COO)_2(aq) + 4.5H_2O + 0.75O_2(g) \end{gathered} \tag{5.4}$$

As observed from the results, acetic acid delamination retains the highest level of lithium in comparison to the other organic acids, and this grants it the highest first-cycle capacity of 84.5 mAh/g. All the acids delaminate the electrode well, with close to 100% efficiency in separation, apart from malic acid, which only has 86% efficiency in separation but leaches only low levels of transition metals and Al into the acidic solution. As shown in Figure 5.2 and Table 5.3, the X-ray diffraction patterns do not significantly change, and LMO and NCA are apparent in all the samples. However, the proportion of the two phases changes, deviating from the original 3:1. This is attributed to transition metal leaching by the acids. It was observed in the ICP data (Figure 5.1) that Mn was preferentially leached by the acids. This is similar to what has been found via previous processes [42–45]. This correlates to the large reduction in LMO content in the pimelic and succinic acid samples.

The valorisation of black mass derived from recycled lithium-ion batteries plays a critical role in promoting resource conservation and waste reduction and involves a series of steps to extract valuable materials from the electrode materials [28, 66]. Figure 5.5 schematically shows the process flow for the valorisation and recycling of an end-of-life battery. The substitution or inclusion of the organic acid delamination steps are highlighted in orange.

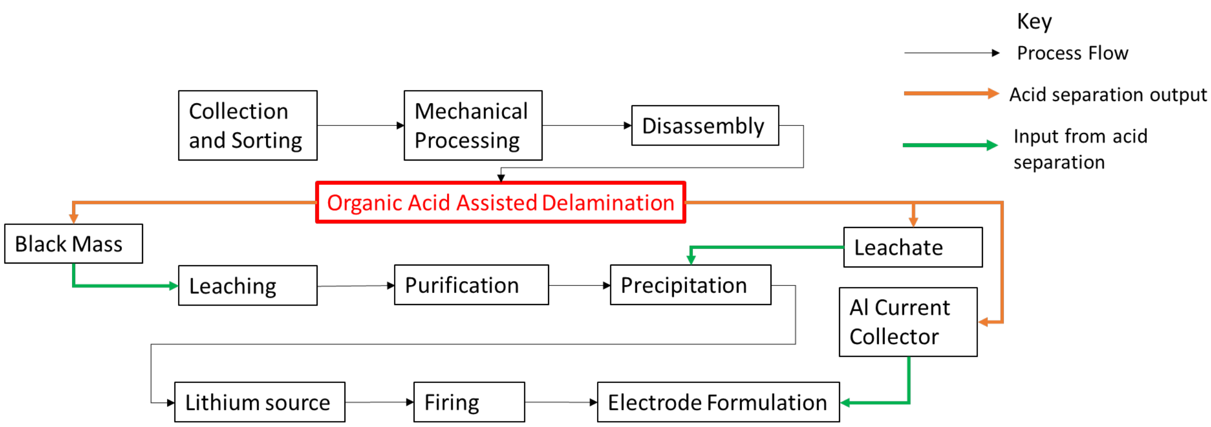


Figure 5.5: Process flow of valorisation and the recycling of an end-of-life battery.

After disassembling or shredding and sorting, the materials are concentrated into their most valuable components. To do this, the electrodes are delaminated from the current collectors to liberate the active material components, and then the binder is negated. For maximum valorisation of the cathodic black mass, a concentration of the transition metals is required, with the aluminium foil or contaminants removed prior to further processing.

This work shows that all organic acids can aid the delamination of the cathode electrode from the current collector. Oxalic and malic acids show the lowest lithium retention, with acetic acid retaining the most lithium. With respect to cobalt, all the acids are within 6 PPM of each other the exception to this is citric acid which retains 65 PPM of cobalt. Acetic, lactic, succinic and pimelic acids all show good retention of manganese with all having over 800 PPM of manganese, citric acid has the lowest retention with 436 PPM of manganese retained in the black mass sample. Acetic, lactic, succinic, pimelic, as well as oxalic acids show good nickel retention with over 400 PPM, again citric acid shows poor retention performance with only 250 PPM. If shredded material streams are considered, it is important to separate out different materials effectively prior to delamination to prevent cross-contamination. Following the black mass concentration, a leaching process to reclaim the lithium and transition metal salts can be performed. these can then be used to re-manufacture cathode active material. Leaching the shredded battery fragments or disassembled electrodes typically involves treating the materials with mineral acids to dissolve and separate the valuable metals from the other components, such as the binder and carbon. Copper is likely an additional contamination in these waste streams.

After leaching, purification of the compounds is required. Depending upon the level of impurities, several different purification steps may be required before the cathode material can be re-manufactured. In these cases, aluminium is the major contaminant from the current collector. This, however, stays in the leachate or solution rather than the reclaimed active material black mass. Therefore, this organic delamination method reduces the number of purification steps required. Short-loop recycled cathode materials are manufactured through the precipitation of metal hydroxides from the precursor salt solutions, intimately mixed with a lithium source and then fired at high temperatures. Directly recycled cathode materials require lithiation with the cathodic black mass, which may require further binder negation before re-lithiation, and the cathode type will be limited to that which was in the end-of-life cell. These cathodes can then be processed back into an electrode and used in a cell. For both direct and short-loop recycling, the elimination of impurities is important. Although outside the scope of this work, an economic and LCA study to determine if direct recycling with a either lithiation step or simply leaching and upscaling would provide more detailed information on the preferred recycling methods (this is outside the scope of this work).

A further benefit of the organic delamination process is zero waste, where in this case waste is defined as any component that is not recovered but has value (e.g. leachate solution containing transition metals), or water that can not be safely discharged into the environment. The organic acids can easily decompose and are biodegradable, aluminium metal is easily reclaimed and recycled, the valuable transition metals contained within the active material are recovered in the black mass as a solid for further processing [160, 161], and although the metal content in the acid leachate is limited, further recovery is possible through nanofiltration and other methods [162, 163]. A zero-waste approach is going to become more and more vital as the demand for critical metals such as Ni and Co increases due to automotive electrification, and the development of techniques to improve recovery rates is essential [164].

This work provides routes to improve the purity of the recycling feedstock from either disassembled or shredded cells. Organic acid delamination removes the cathodic black mass from the current collector with limited leaching of any of the aluminium, lithium, or other transition metals.

5.5 Conclusions

This study presents a scientific overview of the valorisation process, which involves extracting valuable components or materials from waste or by-products. The process aligns closely with the principles of the circular economy, aiming to keep resources in use through recycling, reusing, and recovering materials. The current focus on black mass valorisation lies in the recovery of transition metals. The findings reveal that all acids can extract transition metals and lithium to some extent, but certain acids exhibit selective leaching or lower levels of extraction compared to others. All the acids tested have a black mass recovery of >86% and acetic acid showed 100% black mass recovery. Among the acids tested, acetic acid exhibited the highest lithium retention 66.76 PPM) and the highest first-cycle specific capacity was observed when electrochemically tested (84.51 mAh/g). Depending on what transition metal is desired for retention or leaching different organic acids can be considered the best. If for instance nickel is desired as the transition metal to be retained in the black mass then succinic acid should be used for separation, on the other hand, if cobalt is the targeted metal for retention then citric acid should be used. If first-cycle capacity is the target metric then acetic acid performed best.

Although this work has been able to separate the valuable cathodic black mass from the current collector without fully dissolving the current collector as has been the issue with many acid-based processes, the process still yields some waste containing both lithium and other valuable transition metals, in particular in the acidic leachate. In the next chapter, This work will attempt to mitigate this and head towards a zero-waste process by using a green solvent to negate the ability of the binder to adhere the black mass to the current collector.

Chapter Six

Binder Negation

6.1 Introduction

This work aimed to produce valorised and pure cathodic feedstock from Nissan Leaf Generation 1 cells containing LMO/NCA mixed cathode active material to feed into recycling processes using environmentally friendly processes. In this work, two separate agitation methods were investigated at a range of temperatures separately and concurrently. These were ultrasonic agitation and stirring using an overhead mixer in conjunction with DMSO a green solvent (the benefits and drawbacks of DMSO versus other solvents will be discussed later in this chapter). Once the cathode had undergone the processing steps ICP-OES was performed to assess the transition metal content of the recovered black mass with a particular interest in Ni, Co, and Mn, and the separation efficiency was calculated according to equation 6.1. This work shows that at 60℃ and in only 10 minutes of processing more than 84% of aged material can be removed from the current collector. It has also also been shown that at the same conditions up to 95% of fresh material can be removed from the current collector.

In this work, two types of materials were used: aged material and fresh material.

Aged material in this work is defined as material that has been extracted from cells and left exposed to the atmosphere in storage after washing for a minimum of 4 weeks. This is to approximate the conditions likely to happen if the disassembly and recycling processes are carried out at different locations and it has to be shipped. Fresh material, as referred to in this work, is material that is extracted from cells and used within a week; this is to approximate a process where disassembly and recycling are carried out at the same site and transport is minimal. Another novelty of this study is that it investigates a process using dimethyl sulfoxide (DMSO), which unlike previously investigated solvents [114] is a green solvent that in its pure state is considered non-hazardous and is produced from a by-product of the Kraft process [165]. Also unlike previously reported solvent-based binder negation techniques, it uses low temperatures ($T<65$℃) [114] to negate the binder and reclaim active material. Three approaches were investigated, heating and ultrasonication, heating and stirring, and heating stirring and ultrasonication.

6.2 Methodology

6.2.1 Heating and Ultrasonication

DMSO was heated using a hotplate and the cathode material was added to the DMSO in a 10:1 liquid:solid ratio. This was then placed into a shiesto ultrasonic bath (100 W), and the sample was ultrasonicated for 10 minutes at 50% (50 W) power. This was repeated at 30-80 °C in 10 °C increments. Once the 10 minutes had elapsed, the beaker was removed from the bath, the DMSO was poured off, and the remaining material was washed with two aliquots of 15 mL of room-temperature DMSO. The material was then placed in a single layer on a sample tray and placed into a 60 °C oven to dry overnight.

6.2.2 Heating and Stirring

50 mL of DMSO was measured using a measuring cylinder into a beaker, and the beaker was heated using a hotplate. Once at temperature, the cathode material was added to the DMSO in a 10:1 liquid:solid ratio. This was then placed onto a hotplate with a separate beaker of DMSO with no cathode material. The temperature probe was placed in the DMSO container with no cathode material in order to keep the temperature constant. The stirrer rod powered by an overhead stirrer (Heidolph, Germany) was lowered into the DMSO and cathode mixture and stirred for 10 minutes at 300 RPM. This was repeated at 30-80 °C in 10 °C increments. Experiments were performed in triplicate. After 10 minutes the beaker was removed from the hotplate, the DMSO was poured off and the remaining material was washed with two aliquots of 15 mL of room-temperature DMSO. The material was then placed in a single layer on a sample tray and placed into a 60 °C oven to dry overnight.

6.2.3 Heating, Ultrasonication and Stirring

50 mL of DMSO was measured using a measuring cylinder into a beaker and heated using a hotplate. Cathode material was added to the DMSO in a 10:1 liquid:solid ratio. This was then placed into a shiesto (100 Watt) ultrasonic bath, and a stirrer rod powered by an overhead stirrer (Heidolph Germany) was lowered into the DMSO/Cathode. The samples were stirred concurrently at 300 RPM and ultrasonicated at 50% power for 10 minutes. This was repeated at 30-80 °C in 10 °C increments. The experiments were performed in triplicate. After 10 minutes the beaker was removed from the bath, the DMSO was poured off and the remaining material was washed with two aliquots of 15 mL of room-temperature DMSO. The material was then placed in a single layer on a sample tray and placed into a 60 °C oven to dry overnight.

Once the material had dried, the liberation efficiency was calculated. From previous studies (described in Chapter 4), it had been determined that the cathode comprised 89.8 wt% coating, and the rest of the electrode was the aluminium current collector. Hence the separation efficiency was calculated according to:

$$E_{separation} = \frac{M_s - M_r}{0.898M_s} \times 100 \tag{6.1}$$

where M_s is the starting mass in grams and M_r is the mass of the recovered current collector and any coating remaining on it in grams.

6.3 Results

Figures 6.1 to 6.4 show the comparison of the separation efficiencies of using overhead stirring, ultrasound, and concurrent ultrasound and overhead stirring as agitation methods to liberate black mass from the current collector across a range of temperatures 30-80 ℃. A general trend can be seen in particular from Figures 6.2 - 6.4 where at or above 50 ℃ the increase in separation efficiency starts to slow down with an increase in temperature at 50 ℃ the separation efficiency is 85% and at 80 ℃ the efficiency is 97%, compared to the difference between 40 ℃ and 50 ℃ is 8% and 84%. The error bars in Figure 6.1 40 ℃ ultrasound treated material and 60 ℃ overhead and ultrasound is particularly large due to an outlier result that was included in each calculation.

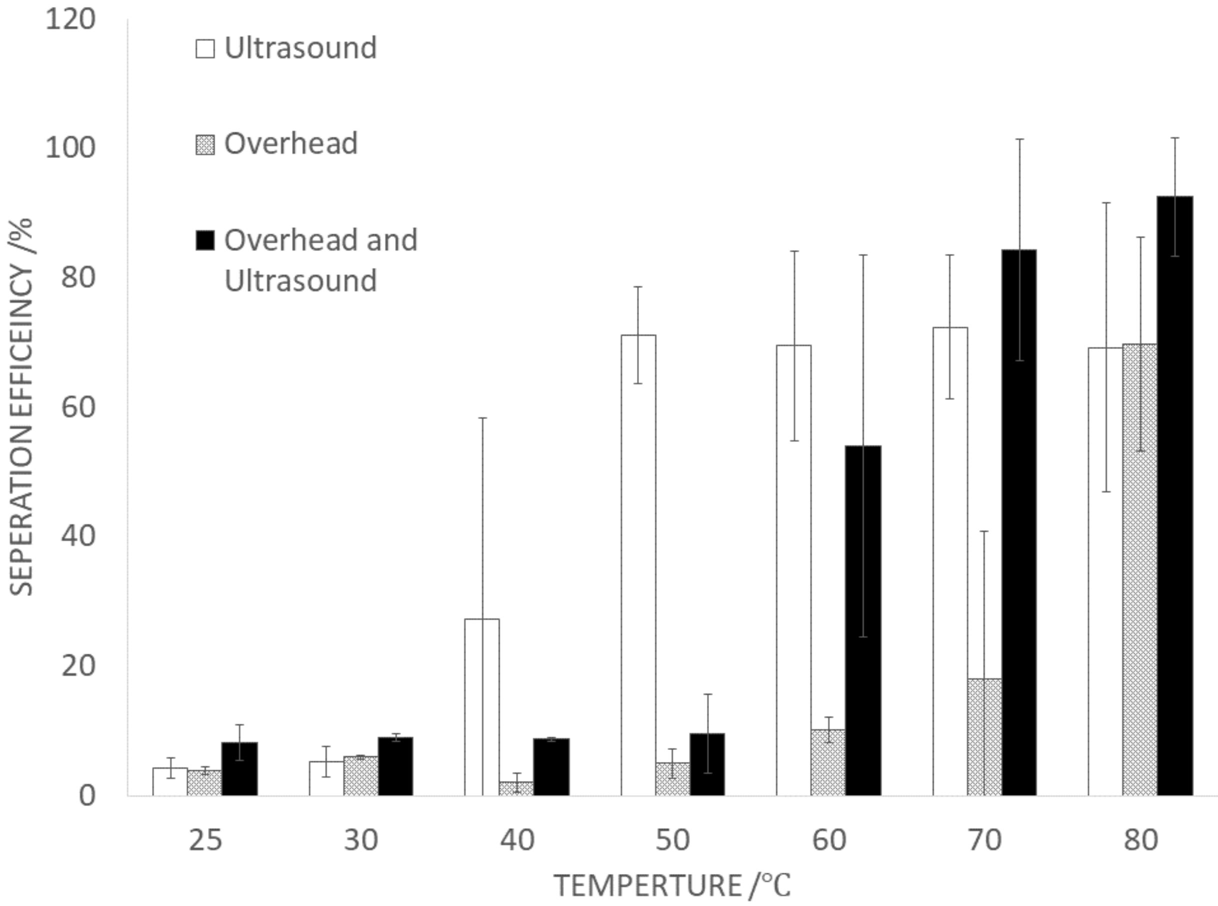


Figure 6.1: Comparison of separation efficiency against processing temperature when using 3 different agitations methods: ultrasound, overhead stirring, and concurrent overhead stirring and ultrasound of aged material processed for 10 minutes.

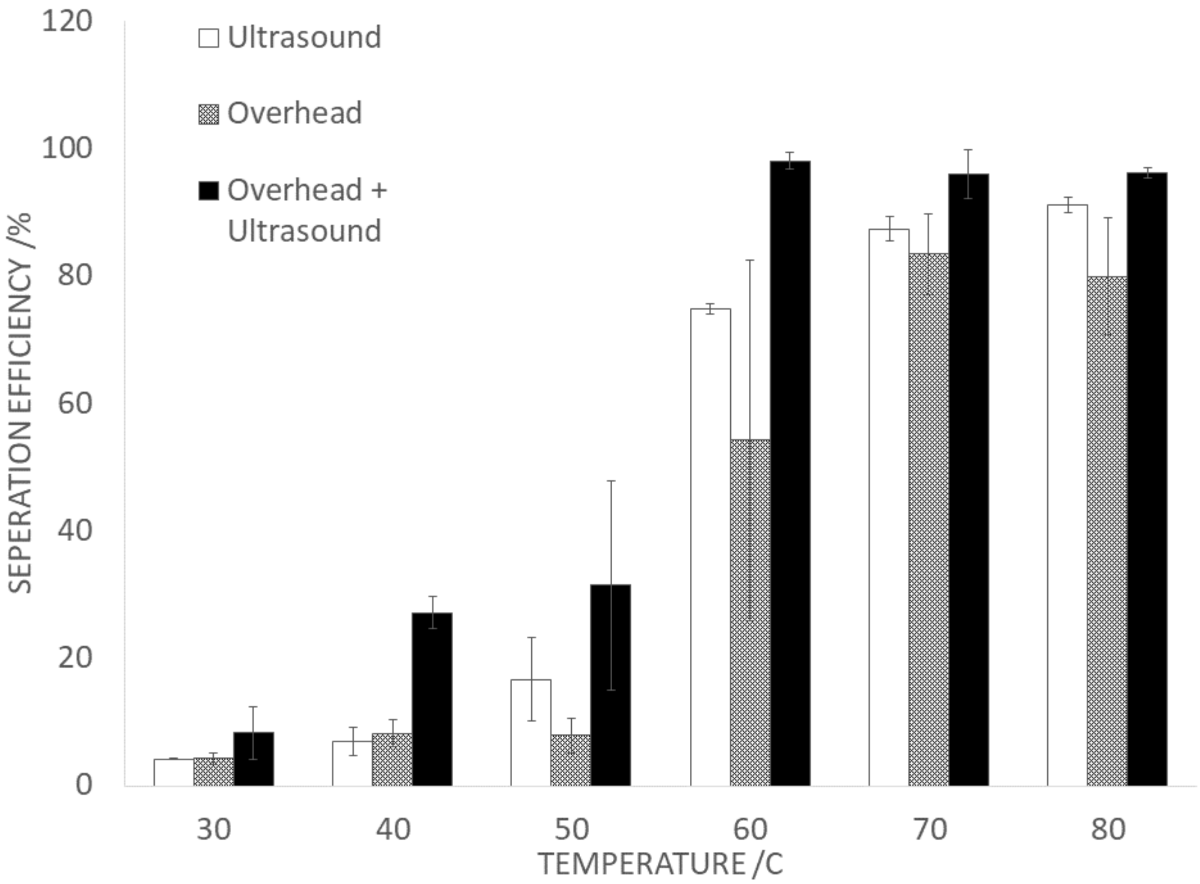


Figure 6.2: Comparison of separation efficiency against processing temperature when using 3 different agitations methods: ultrasound, overhead stirring, and concurrent overhead stirring and ultrasound of aged material processed for 30 minutes.

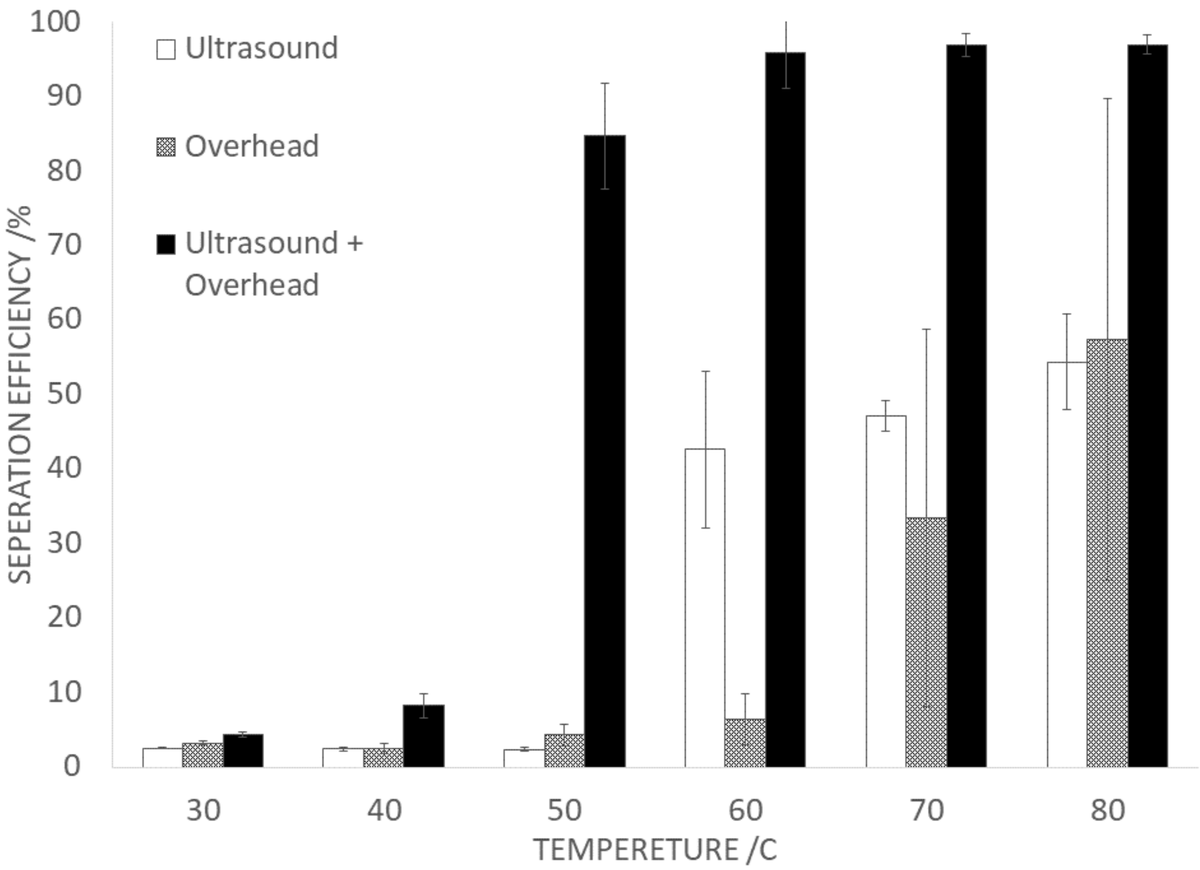


Figure 6.3: Comparison of separation efficiency against processing temperature when using 3 different agitations methods: ultrasound, overhead stirring, and concurrent overhead stirring and ultrasound of fresh material processed for 10 minutes.

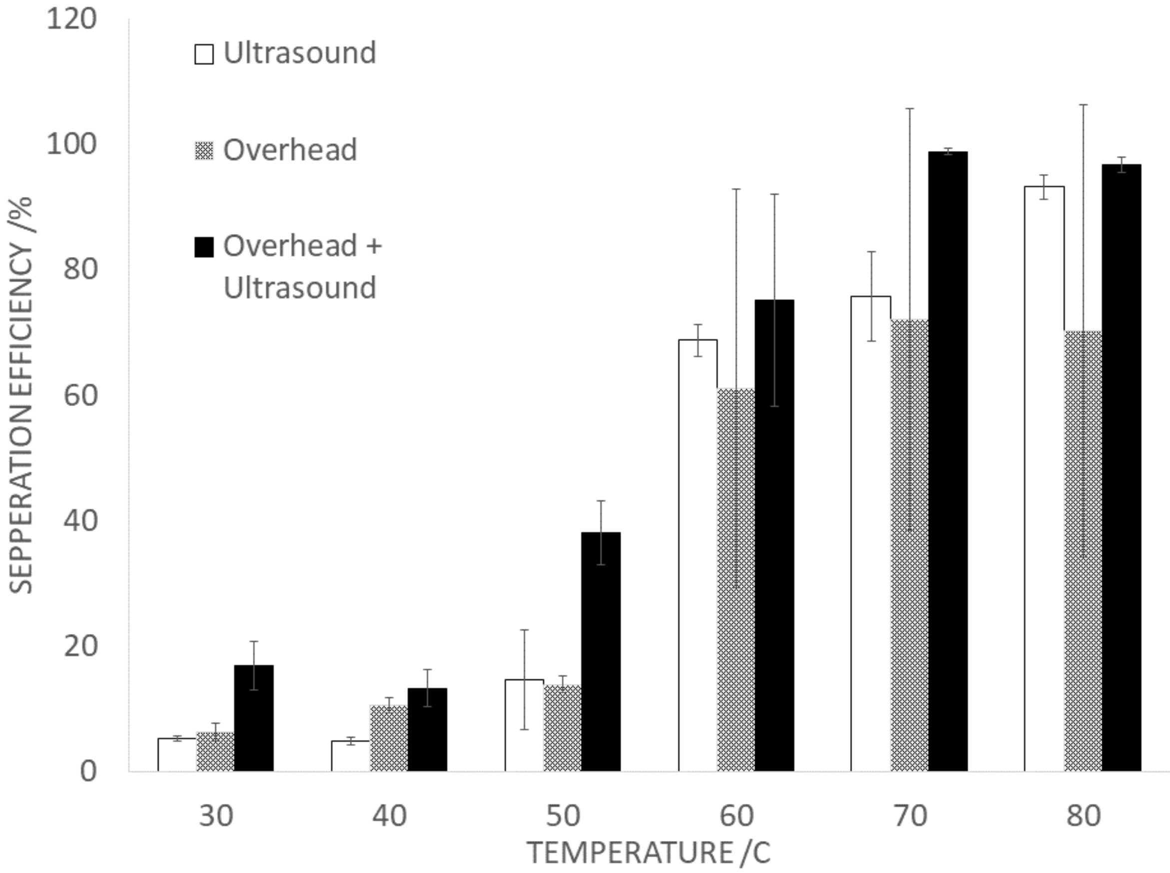


Figure 6.4: Comparison of separation efficiency against processing temperature when using 3 different agitations methods: ultrasound, overhead stirring, and concurrent overhead stirring and ultrasound of fresh material processed for 30 minutes using all agitation methods.

Figure 6.5 shows the results of ICP-OES analysis of metal concentration (Ni, Mn, Co, Li, and Al) from recovered materials and an EoL cell base-line cell on the primary axis and the sample mass of each sample on the secondary axis.

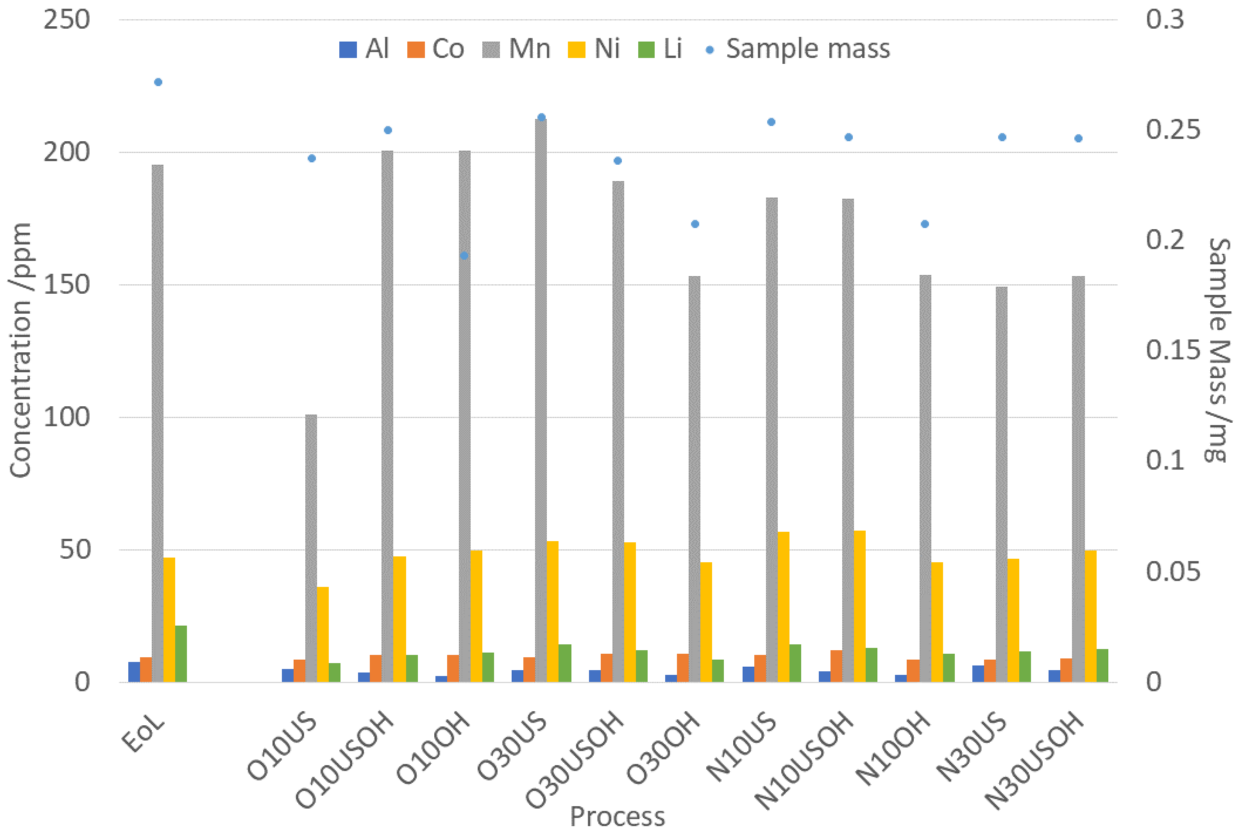


Figure 6.5: Concentrations of Al, Mn, Ni, Li, and Co in DMSO samples and EoL cell on primary axis in PPM. Sample mass of each of the digested samples before digestion on the secondary axis.

Figures 6.6 to 6.15 are the SEM images of the cathodic black matter collected after filtering and drying of the DMSO processed material with Al, Co, Ni, and Mn EDX maps. These particular metals have been chosen for special attention as the cathode consists of a mixed active material LMO:NCA. Mn is the main constituent of the LMO active material and Ni and Co are present in the NCA active material as well as Al however aluminium could also be a major contaminant that would hinder the valorisation of the black mass. An F peak was not prioritised during EDX mapping as there are a number of components that may cause fluorine to be present, these include the binder, electrolyte salt, and potentially degradation products such as LiF. Although these should not be present as they should have been cleaned out in both the washing and recycling steps they do still make an F peak a potentially unreliable way of judging if the binder is still present.

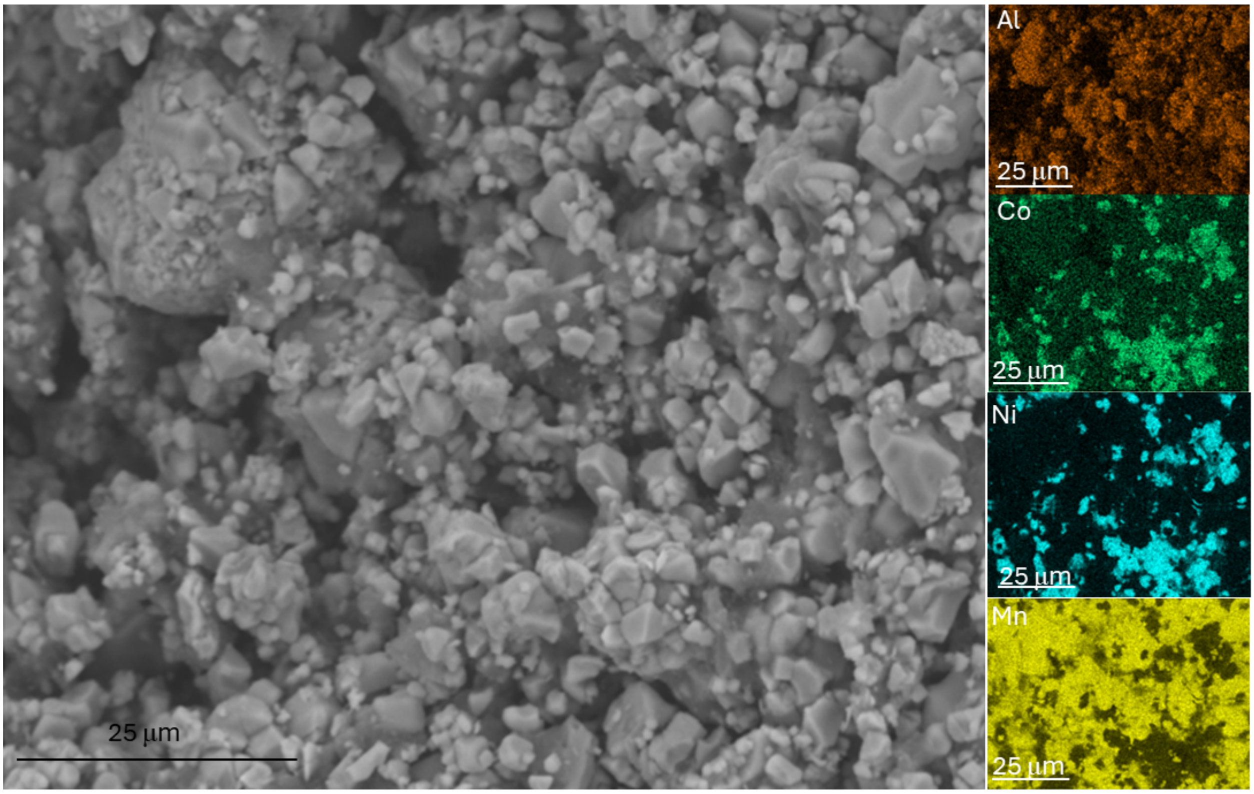


Figure 6.6: SEM image of the collected black mass from fresh material processed for 10 minutes with overhead stirring. Including EDX maps of Al, Ni, Co, and Mn.

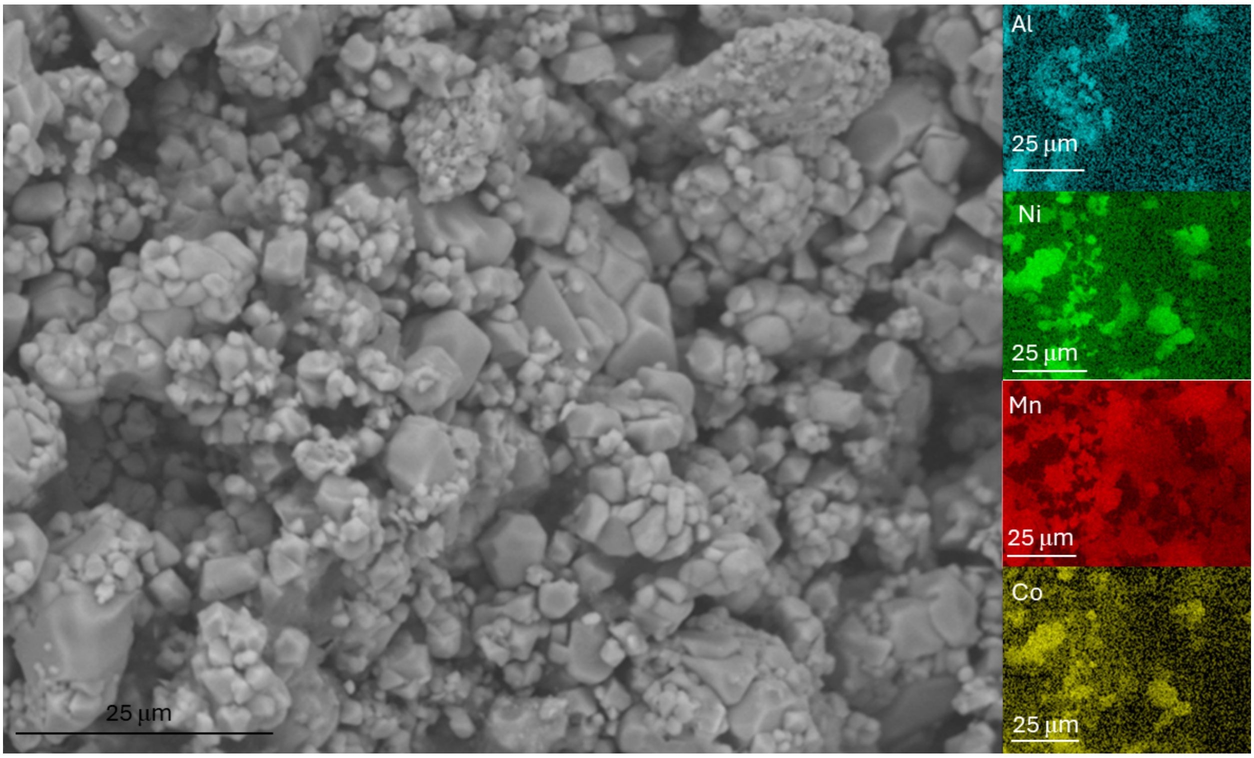


Figure 6.7: SEM image of the collected black mass from fresh material processed for 10 minutes with ultrasound agitation. Including EDX maps of Al, Ni, Co, and Mn.

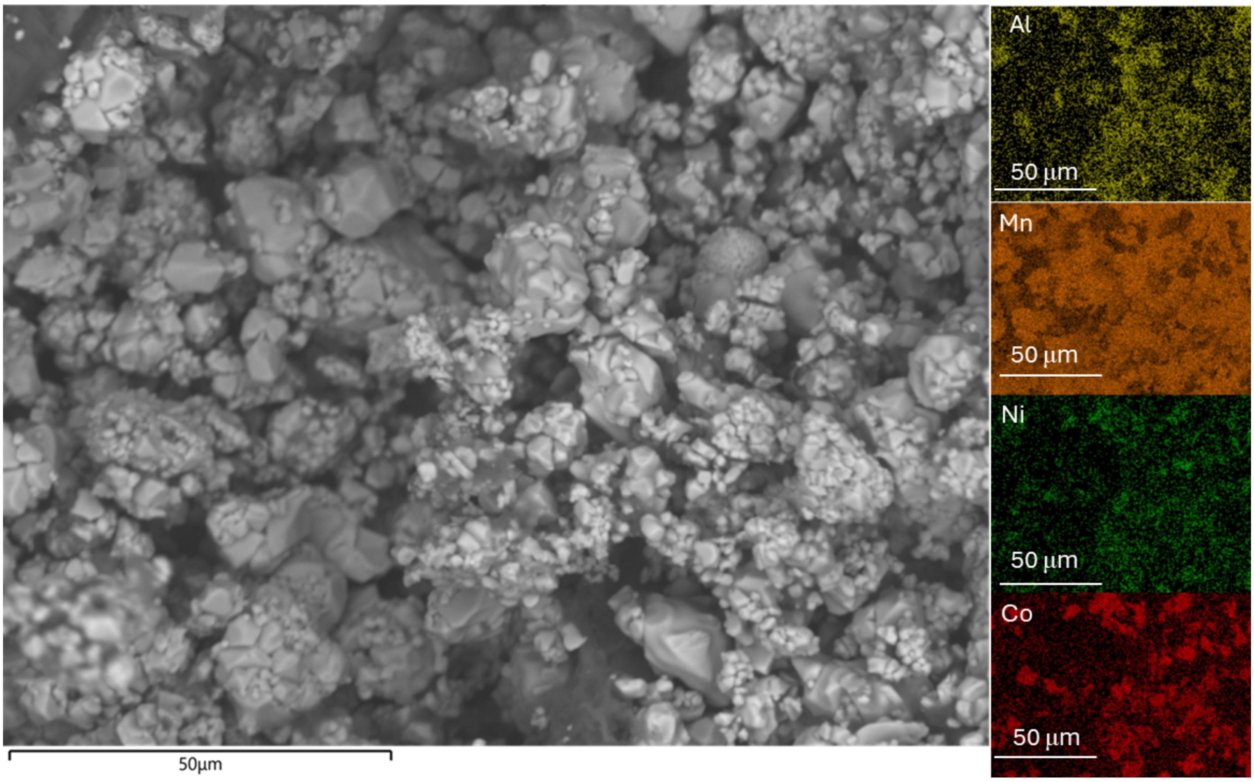


Figure 6.8: SEM image of the collected black mass from fresh material processed for 10 minutes with overhead stirring and ultrasound agitation concurrently. Including EDX maps of Al, Ni, Co, and Mn.

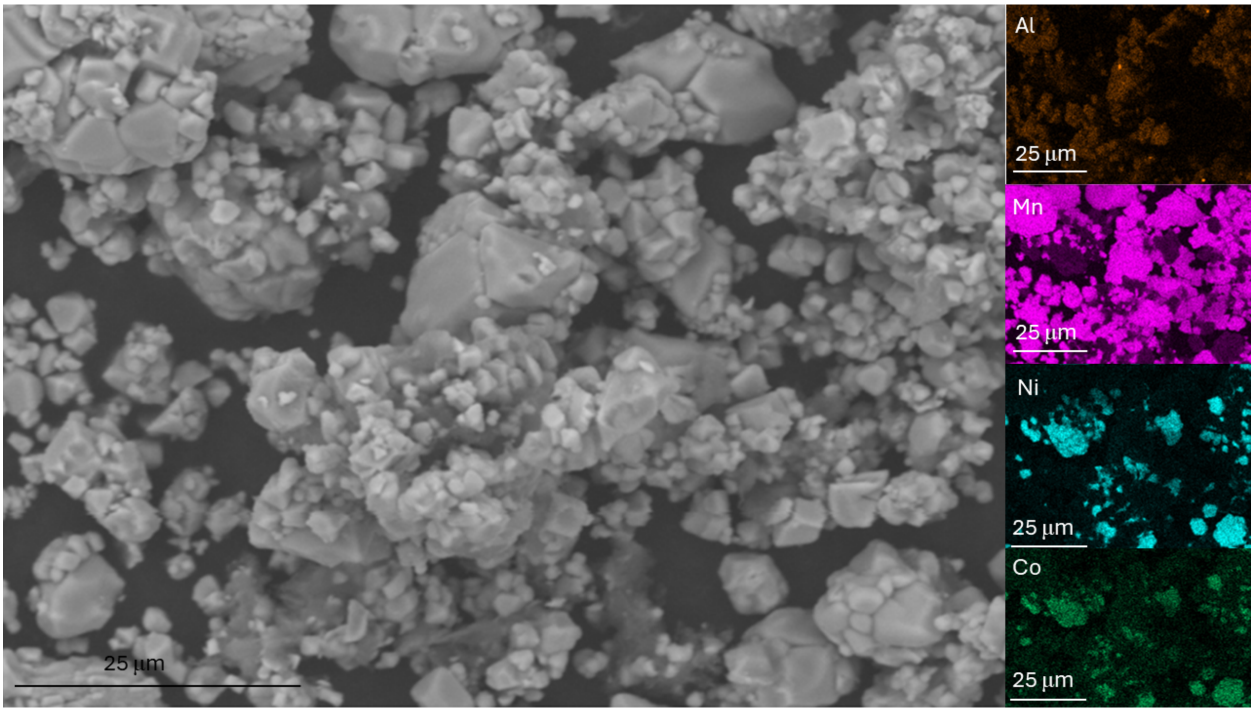


Figure 6.9: SEM image of the collected black mass from fresh material processed for 30 minutes with overhead stirring. Including EDX maps of Al, Ni, Co, and Mn.

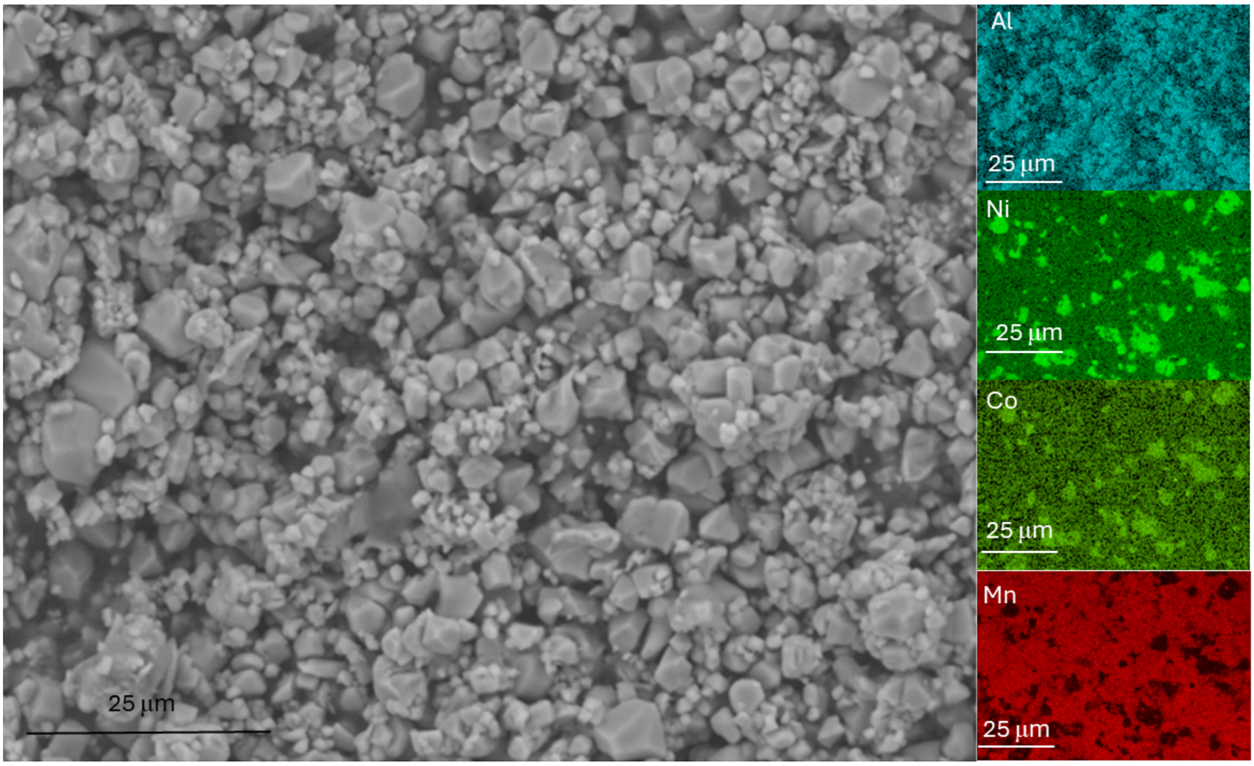


Figure 6.10: SEM image of the collected black mass from fresh material processed for 30 minutes with ultrasound agitation. Including EDX maps of Al, Ni, Co, and Mn.

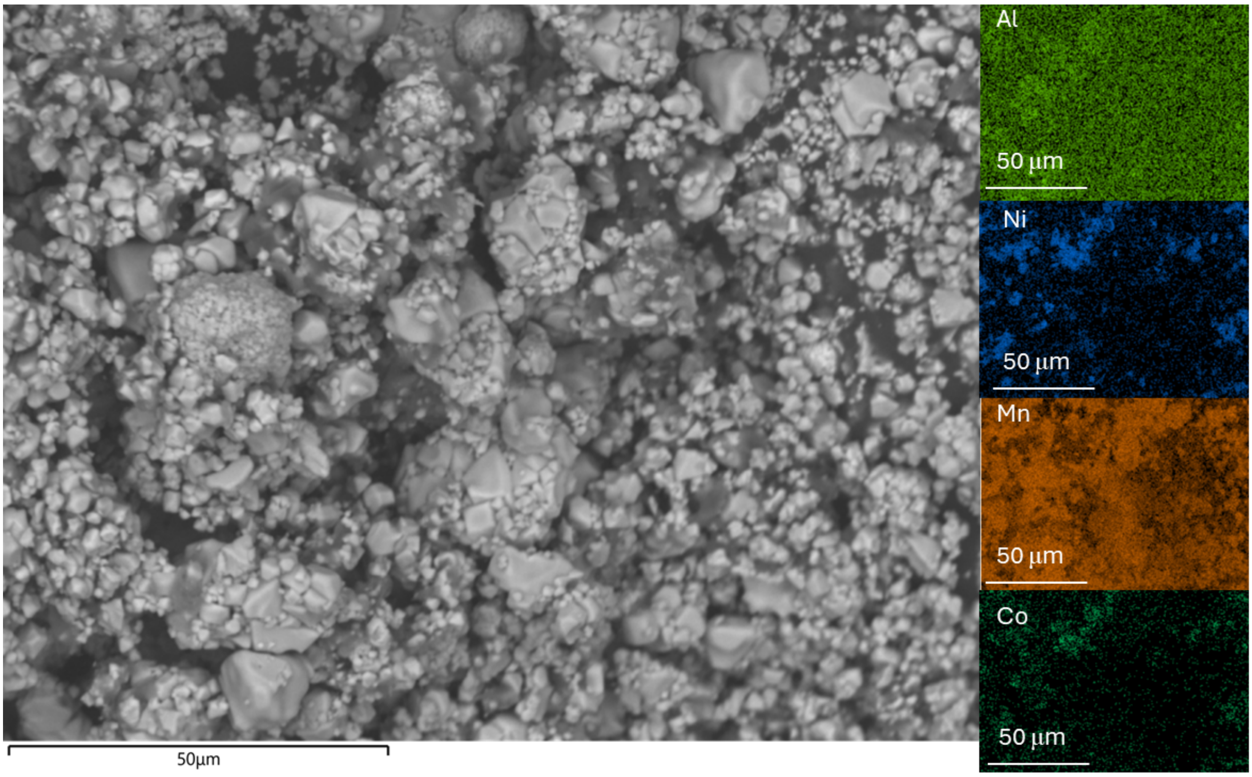


Figure 6.11: SEM image of the collected black mass from aged material processed for 10 minutes with overhead stirring and ultrasound agitation concurrently. Including EDX maps of Al, Ni, Co, and Mn.

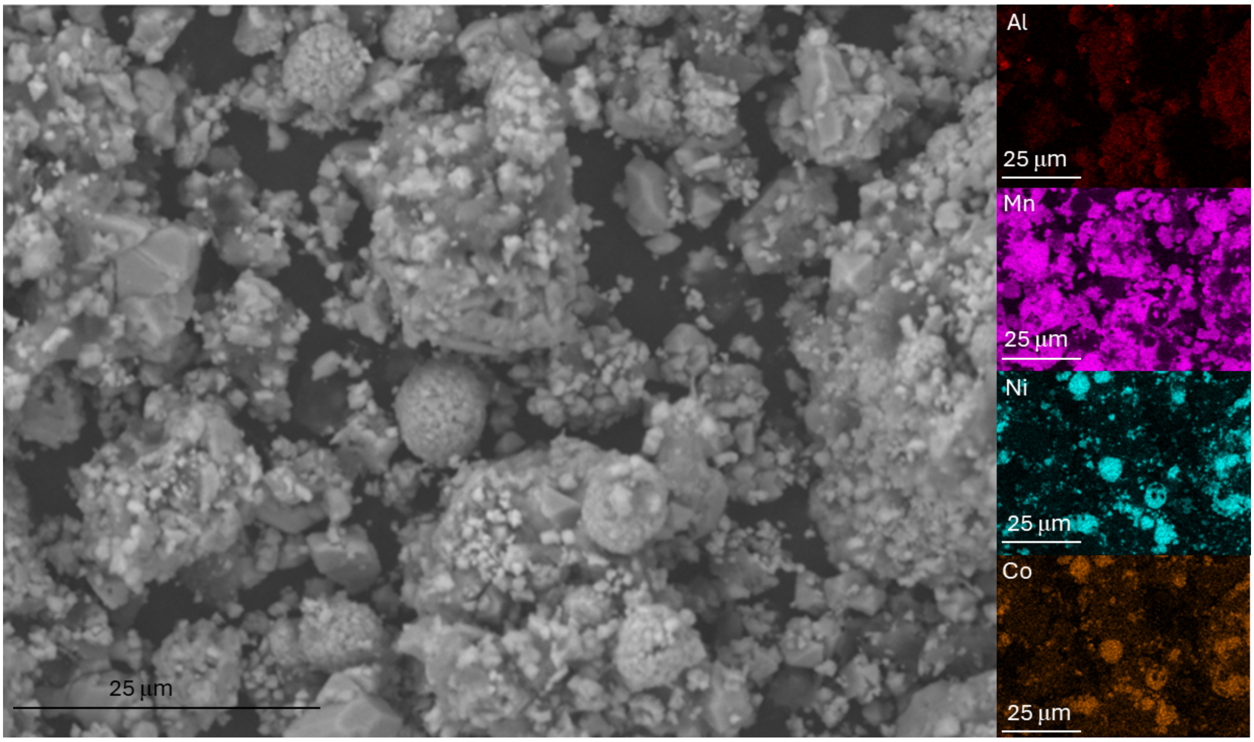


Figure 6.12: SEM image of the collected black mass from aged material processed for 10 minutes with overhead stirring. Including EDX maps of Al, Ni, Co, and Mn.

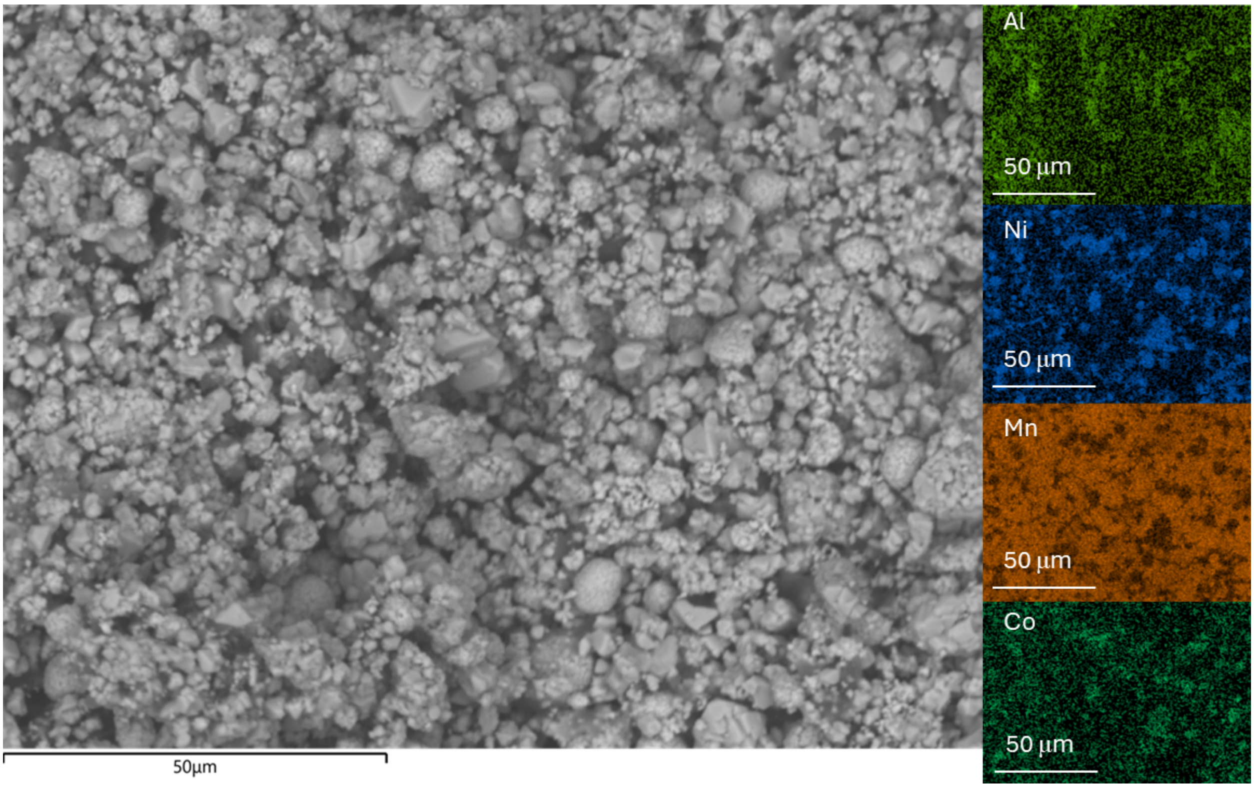


Figure 6.13: SEM image of the collected black mass from aged material processed for 30 minutes with ultrasound agitation. Including EDX maps of Al, Ni, Co, and Mn.

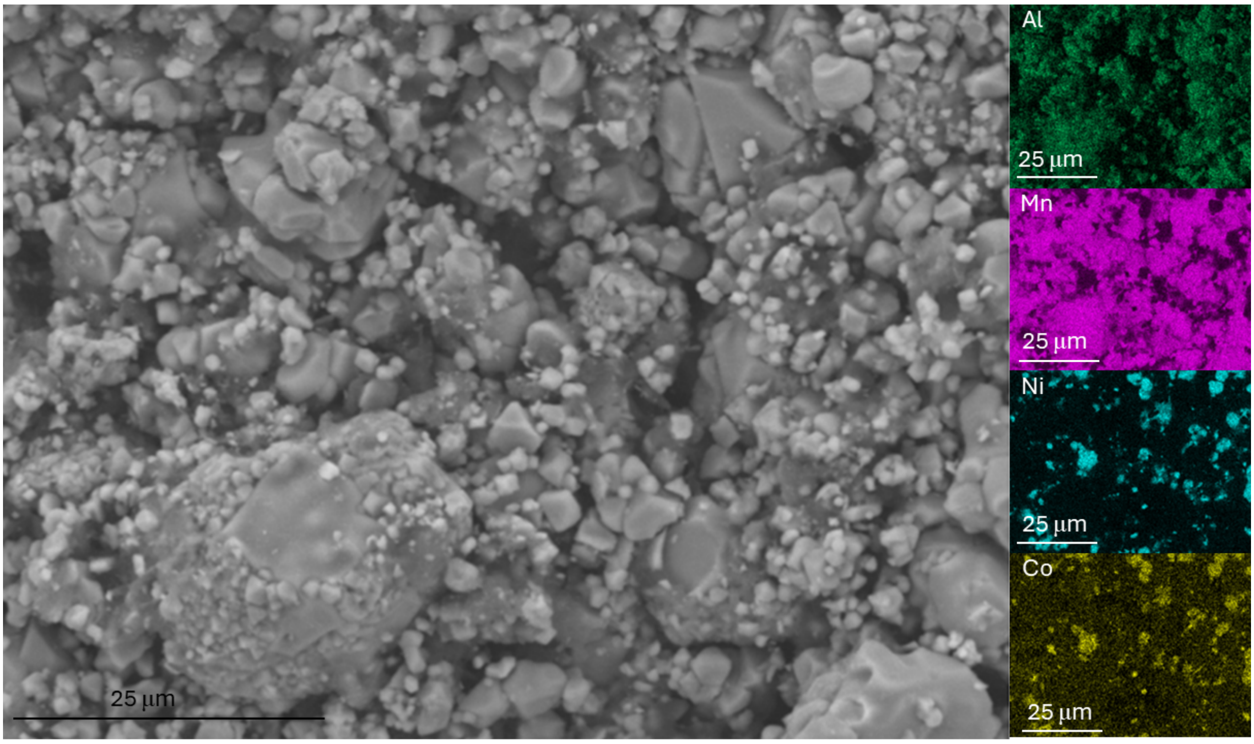


Figure 6.14: SEM image of the collected black mass from aged material processed for 30 minutes with overhead stirring. Including EDX maps of Al, Ni, Co, and Mn.

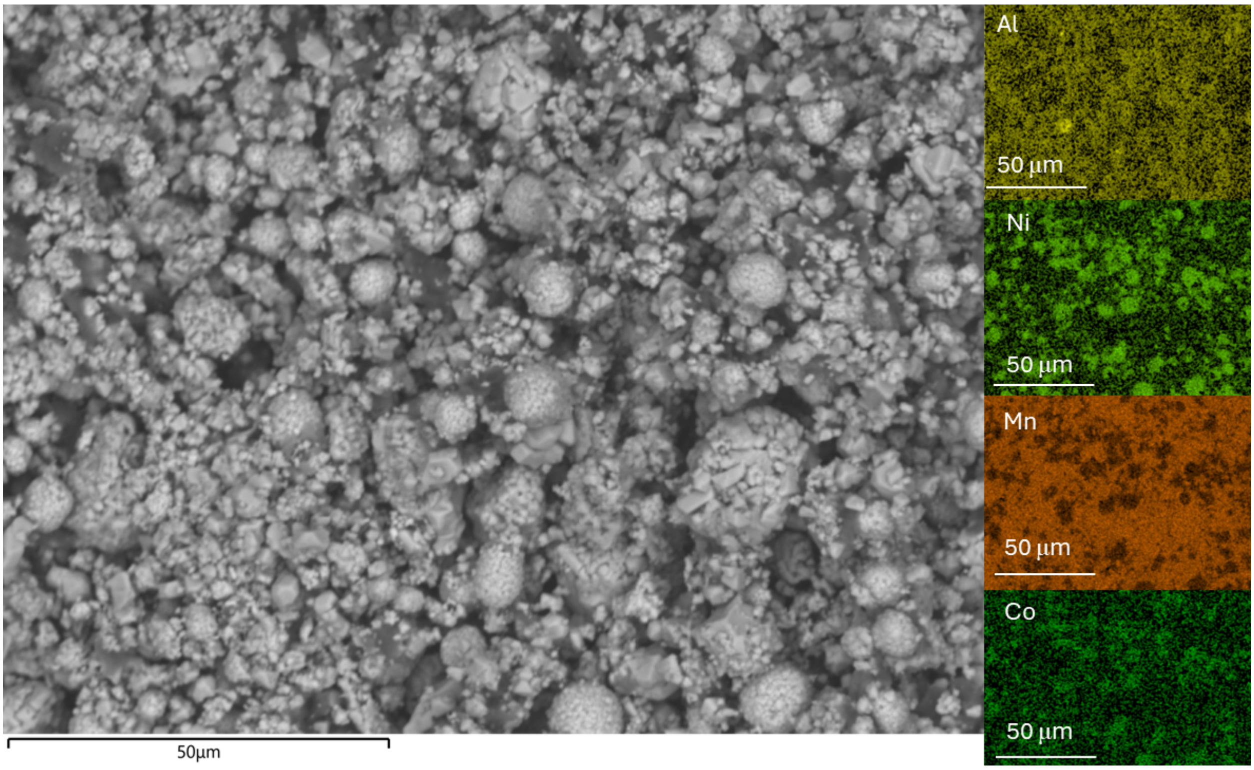


Figure 6.15: SEM image of the collected black mass from aged material processed for 30 minutes with overhead string and ultrasound agitation concurrently. Including EDX maps of Al, Ni, Co, and Mn.

6.4 Discussion

6.4.1 Solubility and Safety

The ability of a solvent to dissolve a polymer can be inferred by using Hansen solubility parameters [166]. As defined by equations 6.2 and 6.3

$$(Ra)^2 = 4(\delta_{d2} - \delta_{d1})^2 + (\delta_{p2} - \delta_{p1})^2 + (\delta_{h2} - \delta_{h1})^2 \tag{6.2}$$

$$RED = \frac{Ra}{R_0} \tag{6.3}$$

Where: Ra is the difference between Hansen solubility parameters for a solvent ($_1$) and a polymer ($_2$); δ_d is the energy from dispersion forces between molecules; δ_p is the energy from dipolar intermolecular forces between molecules; δ_h is the energy from hydrogen bonds between molecules; R_0 is the radius of the interaction sphere in Hansen space; RED stands for relative energy difference.

These were used to produce table 6.1 summarising the ability of some select solvents to dissolve PVDF for which the $\delta_{d/p/h}$ data is included in the top line of the table for the purposes of RED calculation.

Table 6.1: Summary of Hansen solubility parameters of select solvents.

Solvent	δ d	δ p	δ h	Ra	RED
PVDF	17.2	12.5	9.2		
NMP	18.4	12.3	7.2	3.1	0.44
DMSO	18.4	16.4	10.2	4.6	0.66
DMF	17.4	13.7	11.3	2.4	0.35
Ethylene glycol(EG)	17	11	26	16.8	2.3
diethylene glycol (DEG)	16.2	14.7	20.7	11.8	1.6
glycerol	17.4	12.1	29.3	20.1	2.8

Any value of RED$<$1 is considered to be a good solvent that will dissolve the polymer. From table 6.1 as can be seen from the table two non-NMP solvents have a value of RED $<$1, DMSO and DMF. In this work, DMSO rather than DMF was chosen because DMF has a number of hazard statements associated with the pure solvent including flammable, acute toxicity by inhalation and skin contact, eye irritation, and reproductive toxicity. DMSO on the other hand **in its pure form** is considered non-hazardous. However, DMSO is very good at carrying contaminants across the skin barrier and, when a black mass is suspended in DMSO it can carry the heavy metals in the black mass across the skin barrier, and DMSO must be used with latex gloves as it will pass through nitrile gloves.

6.4.2 Mechanisms

Ultrasound works by inducing cavitation bubbles with high-frequency pressure [167]. This cavitation causes the coating to fracture allowing the solvent (DMSO) to penetrate deeper into the coating and dissolve the binder (PVDF) more efficiently due to an increase in surface area. Overhead stirring is a physical process that moves the sample and solvent around, this allows for any liberated black mass to be suspended in the solvent meaning that fresh binder is always exposed to the solvent hence allowing for the dissolution of the binder to occur more efficiently.

Due to the different ways that the two agitation methods act it was thought that combining them concurrently would yield the highest liberation efficiency. This is supported by the Figures 6.2 - 6.4, however, it appears the most important factor in the separation efficiency is the temperature at which the processing occurs as conducting the process at 60 °C or above yields separation efficiencies over 85%. Other studies have used glycerol to separate black mass from current collector [114], however, in that work very high temperatures (over 150 °C) were employed to begin separation and good separation was not achieved, hence although an environmentally acceptable solvent is used the energy consumption of that process is considerably higher.

6.4.3 Material characterisation

In Figure 6.5, it can be seen that there is some variation and loss of metals from the EoL base-base line in the processed materials. This is unexpected as this is a solvent and should not be leaching any transition metals. One explanation for this could be that due to the fact that DMSO is hygroscopic, and as this was done outside a dry room environment and the DMSO was not dried before the studies it has absorbed water from the atmosphere leading to some of the metals being leached out into the solvent. Particularly affected by this are Al, Mn, Ni, and Li. This is surprising as DMSO has been shown as an effective substitute to NMP by Wang et al. [116] in the manufacturing of electrodes, with no detrimental performance when using virgin cathode active material and PVDF dissolved in DMSO. This suggests that direct recycling may have to occur in a dry room environment with dry solvents. Substitution of NMP by DMSO in the manufacturing process has not been attempted as part of this work and it appears that no further attempts of this have been reported in the literature. This could substantially negatively impact this process's viability and environmental credentials, however, this testing has not been carried out as part of this work and could also not be the solution. Although none of the leaching is constructive to direct recycling or valorisation, the loss via leaching is particularly detrimental as has been shown in the previous chapter (Chapter 5) as this is key to the ability of the material being able to hold its original capacity.

From the SEM-EDX it appears that although there is a reduction in Al (from the ICP results), the EDX mapping for the Al shows that there is contamination over the imaged area, with some concentrated co-occurrence with Ni and Co which is to be expected as the cathode material in the Nissan generation one cell is LMO:NCA as shown in Chapter 4. It appears from the SEM-EDX that there is little to no cross-contamination of Ni or cobalt into the LMO particles or of Mn into the NCA particles. As part of this work, DMSO was only studied on the Nissan Gen 1 cell, however, this would also be applicable to the LGM50 cell cathode formulation discussed elsewhere in the thesis as it appears to be PVDF-based with perhaps the peeling efficacy being different. DMSO however is unlikely to be effective with the CALB formulation as that does not appear to be PVDF-based and so the solvent would have to be changed.

6.5 Conclusions

In conclusion, this work has shown how the green solvent DMSO can be used to separate black mass from the current collector thus valorising the product for further recycling. this work has shown that at temperatures above 60 ℃ and in only 10 minutes of processing more than 84% of aged material can be removed from the current collector. It has also been shown that at the same conditions up to 95% of fresh material can be removed from the current collector.

The study presented in this work can be used as a foundation for using solvents to remove binders from cathodes. This carries the benefit of not dissolving the aluminium current collector unlike the work presented in Chapter 5. As discussed in that chapter although it is a love intrinsic value metal it is energy-intensive to extract and recycle in its non-metallic form. Also unlike the work carried out in the previous chapter, this has a much easier path to a zero-waste (i.e. any useful or valuable components are recovered and any wastewater is safe to be discharged into the environment) process as DMSO can be vacuum distilled and reused whereas the acids do have a limit on the number of times they can be used.

Chapter Seven

Conclusions and Further Work

The work presented in this thesis focuses on understanding how we can move towards zero-waste processes. To understand how we recycle, we need to understand the current composition and construction of the different cells, and if we are to improve our materials reclamation yield, if there is any scope for improved design for disassembly. Chapter 4 characterises three cell types: cylindrical, pouch and prismatic, which are representative of the cell types in the current market. A complete cell tear down and materials evaluation is performed with a critique and discussion of how improvements can be made for design for disassembly and recycling. This is the first time that a range of commercial cells have been compared using the same suit of techniques, and their recyclability has been the focus.

In Chapter 5, the optimisation of cathode material recovery for direct recycling is performed using an acid delamination process. The aim was to retain as much value in the reclaimed cathode as possible and reduce impurity levels improving the valorisation. Through the choice of organic acid and process optimisation, lithium and transition metals were retained in the recovered solid and used for direct or short-loop recycling, limiting the metal leaching into the aqueous waste stream. This is the first time this range of organic acids has been used as a valorisation tool rather than a lixiviant, and the material produced by the acids was assessed for direct recycling.

In Chapter 6, this work investigated the use of DMSO, a green solvent to further improve recovery of the cathodic black mass investigated using solvent delamination to enable better valorisation by avoiding leaching if possible. A number of agitation techniques (stirring and ultrasonication) were used to agitate the solution at a range of temperatures to find the optimum processing conditions for the release of black mass from the current collector. This is the first time DMSO has been used in the recovery and valorisation of black mass, and the first time that optimisation has been carried out on this system.

The key conclusions for each chapter are discussed below.

Chapter 4, this work described created a suit of analysis techniques and principles to safely and quantifiably disassemble a cell, and then analyse the components to an elemental level. The process consists of entering the case via incisions into empty spaces in the cell, disassembly, weight analysis, ICP-OES, XRD-EDX, and XRD analysis. This process has been applied to three of the most common automotive cell designs: pouch, cylindrical, and prismatic. This has also been applied to a much more modern prismatic cell with a successful disassembly and to a super-capacitor, showing that at least the disassembly methods have a wide range of applicability to disassembling energy storage devices. This is the first example where the elemental breakdown between three different formats has been compared, which allows further work on life cycle assessment and supply chain analysis for these cell types. Because of the age of some of the cells, the prominence of the materials is unclear, and this needs to change for future cell types being sold in Europe due to the battery passport implications.

Having disassembled three of the most common large-scale commercial cell formats and discussed the benefits and drawbacks of each cell construction, the following suggestions would make for the most recyclable cell:

1. the outer case: A pouch or prismatic cell with a predefined and triggerable failure point,
2. the electrode stack: A roll or a z-fold stack,
3. the electrode coating: Water-based (or benign solvent) binders are suggested, which contain as few additives as possible that have at least one distinct physical property such as hydrophobicity or distinctly different density to aid in separation.

Having understood what is in a cell and its composition, the recycling process can begin. It will now be possible to assess the efficacy of these processes, track any waste that may be occurring, and understand where value is being retained.

Chapter 5 of this work presents a scientific overview of the valorisation process, which involves extracting valuable components or materials from waste or by-products. The process aligns closely with the principles of the circular economy, aiming to keep resources in use through recycling, reusing, and recovering materials. The current focus on black mass valorisation lies in the recovery of transition metals. The findings reveal that all acids can extract transition metals and lithium to some extent, but certain acids exhibit selective leaching or lower levels of extraction compared to others. All the acids tested have a black mass recovery of >86% and acetic acid showed 100% black mass recovery. Among the acids tested, acetic acid exhibited the highest lithium retention 66.76 PPM) and the highest first-cycle specific capacity was observed when electrochemically tested (84.51 mAh/g). Depending on what transition metal is desired for retention or leaching different organic acids can be considered the best. If for instance nickel is desired as the transition metal to be retained in the black mass then succinic acid should be used for separation, on the other hand, if cobalt is the targeted metal for retention then citric acid should be used. If first-cycle capacity is the target metric then acetic acid performed best.

This work has separated the valuable cathodic black mass from the current collector without dissolving the current collector, as has been the issue with many acid-based processes. The process still yields some waste containing lithium and other valuable transition metals, particularly in the acidic leachate. There is an opportunity for future work to attempt to mitigate this and head towards a zero-waste process by using a green solvent to negate the ability of the binder to adhere the black mass to the current collector.

Chapter 6, of this work illustrates how the green solvent DMSO can separate black mass from the current collector, thus valorising the product for further recycling. I have shown that at temperatures above 60℃ and in only 10 minutes of processing, more than 84% of aged material can be removed from the current collector. It has also been shown that under the same conditions up to 95% of fresh material can be removed from the current collector.

Although much work has been done here to understand cell construction and disassembly and to better valorise material so that more efficient recycling techniques can be developed, much work remains.

Further work could occur in three key areas: design for disassembly, improved valorisation, and waste reduction from recycling processes. Further work for design for disassembly would be to construct a cell that demonstrates the recyclability and separation principles. That is a cell with an easy-to-enter casing and water-based binders, and the active materials are simple to separate from other additives. For improved valorisation, further work could include extensive electrochemical testing to asses the solvent-recovered material and, if needed, the re-lithiation of the reclaimed black mass to investigate the possibility of direct recycling. For the acid-based recovery process, it would be beneficial to investigate if this process can be used as an improved valorisation step for up-scaling unfavourable cathode chemistries into more appropriate ones. This would also reduce waste from this process.

The investigation of turning the acid- and solvent-based processes into zero-waste processes would be beneficial from a waste reduction perspective. The acid process would involve the extraction of transition metals and lithium from the leachate and investigating if the recovery of the acid is at all possible. For the solvent-based process, this would consist of proving that the solvent is extractable and reusable. Also, as the solvent is likely to contain a binder, investigating if this binder is reusable. Another pathway of reducing waste in the solvent will be to investigate if it is possible to directly remanufacture or if needed relithiate and re-manufacture in the solvent thus reducing the volume of solvent used and potentially reducing the number of processing steps (such as the removal of preliminary drying step).

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