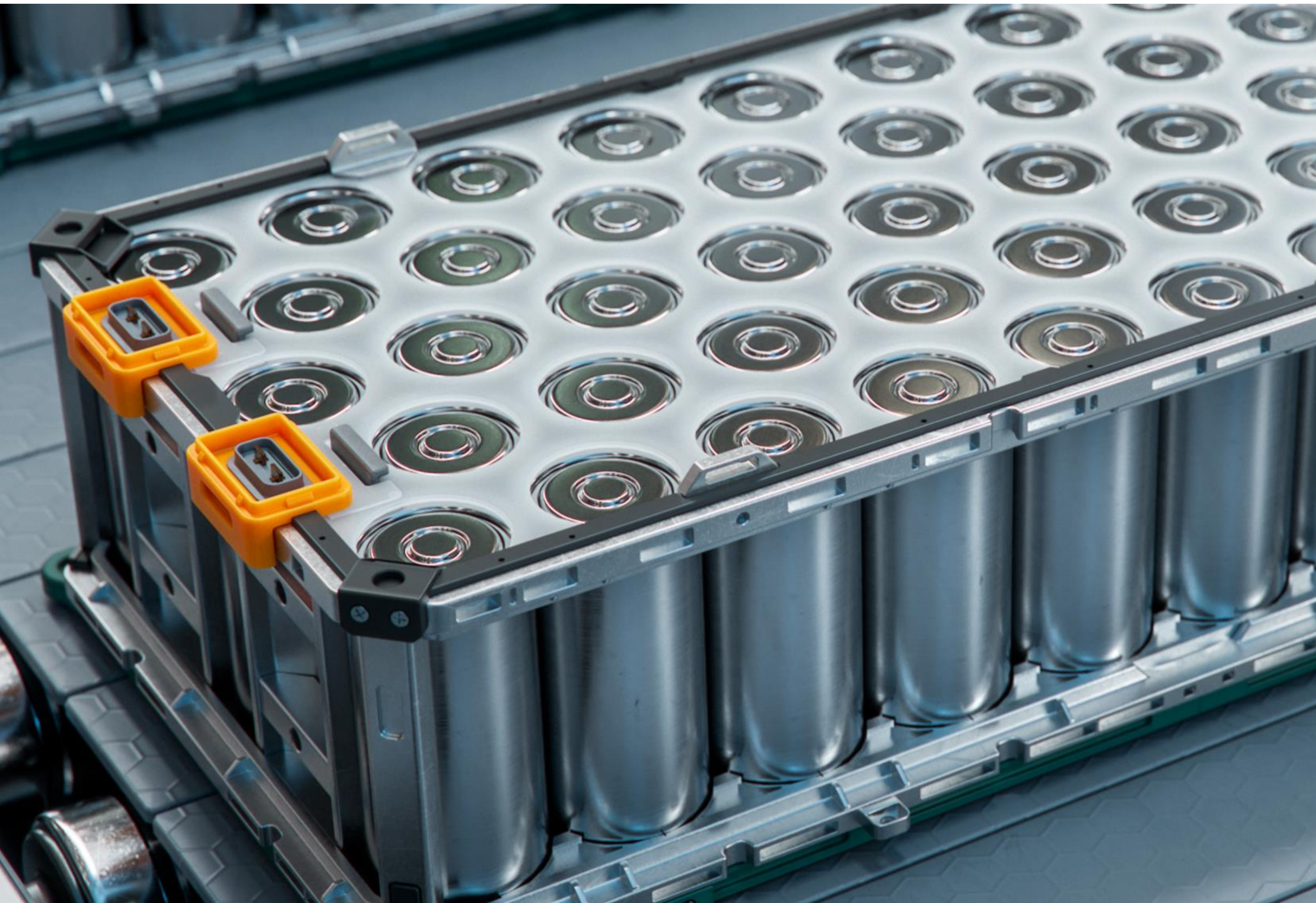
Australia's National
Science Agency

From minerals to materials

Supplementary report: Lithium-ion battery recycling

May 2024



Citation

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Acknowledgements

CSIRO acknowledges the Traditional Owners of the lands that we live and work on across Australia and pays its respect to Elders past and present.

The project team would like to acknowledge the contributions of all stakeholders that provided input to this project from industry, government and academia. Appendix A of the main report 'From minerals to materials: Assessment of Australia's critical mineral mid-stream processing capabilities' includes a complete list of the organisations that provided input into to this project.

Contents

Glossary

Abbreviations

CAM	Cathode active material
EV	Electric vehicle
LCO	Lithium cobalt oxide
LFP	Lithium iron phosphate
LiF	Lithium fluoride
LiPF6	Lithium hexafluorophosphate
NCA	Nickel cobalt aluminium oxide
NMC	Nickel manganese cobalt oxide
NMP	N-methyl-2-pyrrolidone
pCAM	Precursor cathode active material
RD&D	Research, development and demonstration
SEI	Solid electrolyte interface

1 Executive summary

The recovery of high value metals, such as cobalt and lithium, has been the economic driver for commercial activity and research, development and demonstration (RD&D) in lithium-ion battery (LIB) recycling to date. However, supply chain concentration for critical energy minerals and international circular economy policies are driving global efforts to recover more materials from LIB waste, including lithium electrolytes and graphite. Australia's LIB waste is estimated to grow to 136,000 tonnes by 2036, with an associated recoverable value of up to $3 billion. [1]

Australia's RD&D capabilities can support the expansion of LIB recycling activities onshore and increase the value recovered from end-of-life waste streams. There are several opportunities for RD&D related to LIB recycling, including supporting the implementation of mature technologies from overseas at commercial scale; demonstrating Australian IP at scale; accelerating emerging technologies to grow Australian IP; and building new capability in emerging technologies. (Figure 1 and Figure 2).

This supplementary report is part of the report series *From minerals to materials: Assessment of Australia's critical mineral mid-stream processing capabilities*. The series adds to existing Australian and international literature on critical minerals and renewable energy technologies by providing a detailed picture into mid-stream processing, key areas for global risk reduction and capability development to support the energy transition in Australia.

Figure 1: Framework for assessing research, development and demonstration (RD&D) and international engagement actions.

Opportunity area	Establish new capability in emerging technologies	Accelerate emerging technologies and grow Australian IP	Pilot and scale up Australian IP	Support commercial deployment of mature technologies
RD&D actions	Build capability in emerging technology areas via fundamental and applied research projects.	Leverage Australia's strengths to progress technologies beyond the lab and grow Australian IP.	Deploy Australian IP in pilot-scale and commercial-scale demonstrations.	Support the deployment of mature technologies domestically at commercial scale, through commercial testing and validation, and cross-cutting RD&D.
International engagement actions	Engage with research institutions on capability building and knowledge sharing (e.g. joint research programs).	Partner with overseas industry, research or government on mutually beneficial sustained technology development efforts (e.g. co-funded or joint projects).	Engage with upstream offtakers to de-risk and finance pilot projects. Alternatively, demonstrate Australian technologies overseas.	Engage on commercial arrangements e.g. international technology providers, license overseas patents, attract foreign direct investment, and secure offtake agreements.

IP, intellectual property. For a full description and methodology of this framework, refer to the main report *From minerals to materials: Assessment of Australia's critical mineral mid-stream processing capabilities.*

[1] The $3 billion figure represents the lost/recoverable value from LiB wastes in 2036, based on 2021 commodity price. Zhao et al. (2021) Australian landscape for lithium-ion battery recycling and reuse in 2020 - current status, gap analysis and industry perspectives. CSIRO and FBI CRC; King S and Boxall NJ (2019) Lithium battery recycling in Australia: defining the status and identifying opportunities for the development of a new industry. Journal of Cleaner Production 215, 1279–1287. DOI: 10.1016/j.jclepro.2019.01.178.

Discharging

Discharging technology will become increasingly important as EV uptake increases, and as LIB waste poses increasing safety risks across Australia's recycling system. Given the availability of off the shelf discharging equipment, there is an opportunity for Australia to implement discharging systems across the LIB recycling industry, while addressing key barriers such as standards and regulations.

Australia currently undertakes the initial processing steps for LIB waste (crushing and shredding) prior to shipping black mass. Without discharging this can be an unsafe activity for workers and pose a risk to facilities. Given this activity is likely to grow there is a need to state of the art discharging practices within Australia's recycling infrastructure; systems that are safe and do not contaminate materials for subsequent recovery

RD&D can support the development of regulations and standards that currently make electrical discharging (the safest discharging option, and compatible with high quality material recovery) difficult. Further, RD&D in adjacent fields such as robotics and automation can help overcome issues with handling and throughput.

Dismantling and separation

There is an opportunity to improve upon existing separation technologies used in Australian battery crushing and shredding operations. This can increase the quality and price of black mass product and facilitate improved extraction of materials in downstream processing.

Given Australia's current commercial and patent activity, there is an opportunity to demonstrate Australian IP in separation technology onshore, and to continue to drive reductions in cost, energy, and reagent use while increasing revenues from the export of black mass.

Although separation technology is mature and available for industry, improved separation technologies can enable more efficient recovery of high value metals through hydro-metallurgical processes (the subsequent step in processing), as well as the recovery of electrolytes and graphite, which is not currently practiced at scale globally.

Graphite regeneration; Electrolyte recovery

Graphite regeneration and electrolyte recovery is emerging and gaining interest globally; Australia can leverage its RD&D leadership in this area to drive greater circularity and value recovery in domestic and global battery supply chains.

Global battery recyclers are yet to adopt graphite regeneration (as opposed to downcycling) and electrolyte recovery. Given the emergence of this technology and Australia's RD&D activity in this space, there is an opportunity to build on existing capabilities to develop the technology beyond the laboratory and to grow domestic IP.

Pyro- and hydrometallurgical recovery; Electrochemical recovery

The recovery of high value metals using hydro- and pyrometallurgical techniques is commercial overseas, and global RD&D momentum continues to improve upon this technology. Given Australia's patent activity in this space, there is an opportunity to demonstrate Australian IP overseas or alongside emerging domestic battery plants.

Australia does not currently undertake high value metal recovery and instead ships black mass for processing offshore. Despite projected growth, the scale of Australia's LIB waste stream is currently small and geographical dispersion results in challenging economics. Australia's IP in hydro- and pyro-metallurgical recovery can be demonstrated overseas or alongside eventual domestic CAM and battery cell manufacturing plants.

Alternatively, Australia may choose to implement commercially proven technologies from overseas to manage investment risk and accelerate implementation timelines. RD&D in supporting fields may be beneficial, such as assessing the economic viability of overseas technologies in the Australian context.

Direct recycling; Biohydrometallurgy

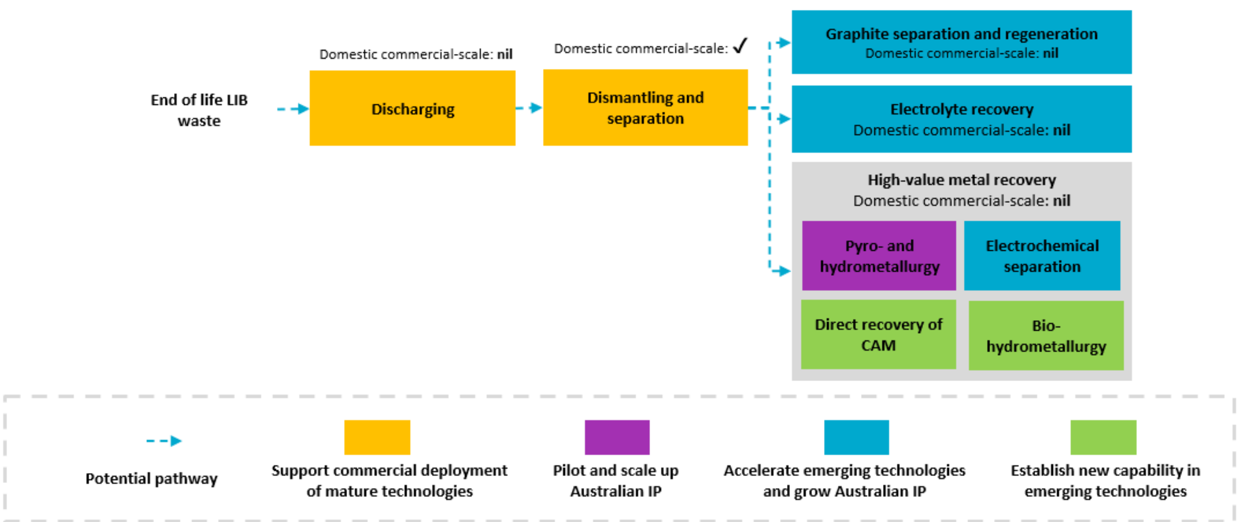
Direct recycling of cathode materials and biohydrometallurgy are emerging technologies that can provide step change improvements in battery recycling. Capabilities are nascent globally and in Australia and can be built through RD&D international collaboration.

Direct recovery of CAM has the potential to reduce the number of steps, inputs and costs compared with commercial processing that targets separated metals. Where applicable, bio-metallurgy has the potential to reduce capital costs, energy consumption and environmental impact.

However, these technologies are at low maturity and several technical challenges will need to be resolved through RD&D to deliver these outcomes.

Australia and partner countries can grow their capabilities in this space through RD&D and engaging in knowledge sharing and joint projects.

Figure 2: Australian RD&D opportunities across LIB recycling technologies.



CAM, cathode active material; IP, intellectual property.

2 Objectives and scope

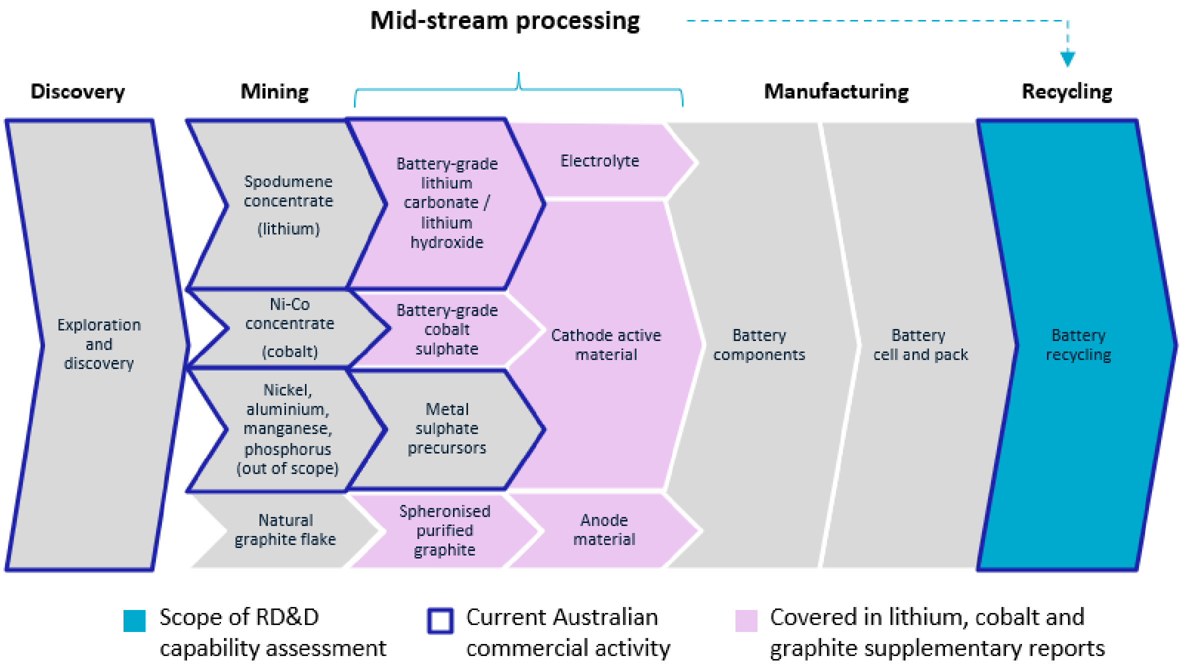
This supplementary report focuses on Australia's key lithium-ion battery (LIB) recycling gaps, and aims to address several objectives:

- To introduce the key current and emerging technologies underpinning the processing of LIB waste, with a focus on the production of battery grade materials to re-introduce into the battery supply chain.
- To present the level of IP and research activity occurring in Australia and globally, for each emerging and mature technology area.
- To identify key challenges and opportunities for Australia to build domestic IP and collaborate with international partners, based on technological maturity, IP trends and research activity.

The importance of reusing second life batteries should not be overlooked, however is not included in this report chapter due to its focus on processing technologies.

The purpose of this analysis is to guide and inform government, industry and research decision-making with respect to research, development and demonstration (RD&D) investment and collaboration efforts across critical minerals and renewable energy technology supply chain activity.

Figure 3: Scope of LiB recycling supplementary report and current commercial production in Australia.



Ni, Nickel; Co, Cobalt.

3 RD&D challenges and opportunities

In light of increasing demand for LIBs, Australia has the opportunity to establish a circular economy by developing a battery recycling industry. Recycling can support Australia's energy transition goals by recovering critical materials to address supply chain risks; provide an economically viable revenue stream; provide a safe solution for the disposal of dangerous LIB waste; and mitigate environmental risks of disposal to landfill.

The recycling of used batteries comprises multiple stages, some currently undertaken by industry, and others more emerging with potential for greater resource circularity (see Figure 4).

This section will discuss the RD&D challenges and opportunities relating to mature and emerging technologies for recycling LIB batteries.

- Section 3.1 will cover technologies used for discharging end of life LIB. The lithium battery recycling process begins with the discharging of collected batteries to ensure safety during transport, storage, and further processing. This mitigates fire risk and electrical risks to personnel stemming from the residual energy retained within batteries.[2]
- Section 3.2 will cover dismantling and separation processes, which sometimes involves a deactivation step. This prepares battery waste for metallurgical processing, with the goal of isolating the internal materials that contain the target metals to be recycled.[3] Shredded mixed material is also known as 'black mass'.
- Sections 3.3 and 3.4 will cover graphite regeneration and electrolyte recovery. These are emerging processes targeting recycling of graphite into battery grade anode material, and recovery of valuable electrolyte components such as lithium. This begins at the separation stage, followed by further processing.
- Section 3.5 will cover high value metal recovery, targeting the extraction of materials, such as lithium compounds, cobalt and nickel. These processes typically use pyrometallurgy, hydrometallurgy, or both.[4] Emerging methods to directly recover and regenerate the cathode active materials (CAMs) are also being explored due to the reduced costs and energy requirement compared to extracting individual metals. Similarly, bio-hydrometallurgical extraction is an emerging low-cost alternative to these methods and is undergoing further research.

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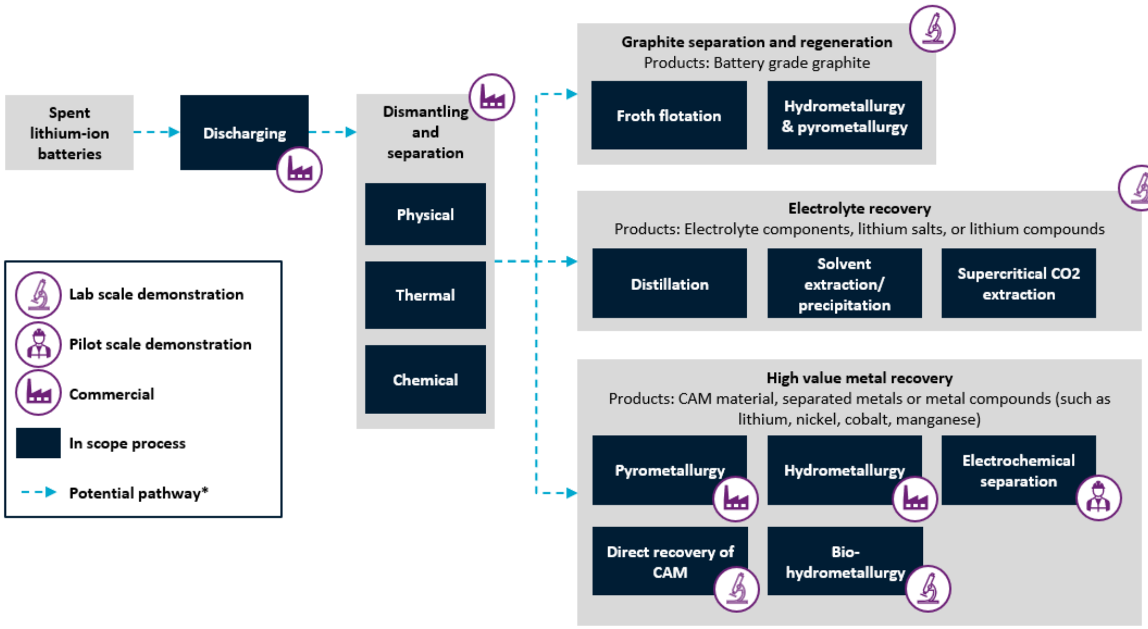
[2] Zhao et al. (2021) Australian landscape for lithium-ion battery recycling and reuse in 2020 - current status, gap analysis and industry perspectives. CSIRO and FBI CRC.

[3] Espinosa R, Mansur B (2019) Recycling batteries. In Waste Electrical and Electronic Equipment (WEEE) Handbook. Woodhead Publishing.

[4] Espinosa R, Mansur B (2019) Recycling batteries. In Waste Electrical and Electronic Equipment (WEEE) Handbook. Woodhead Publishing.

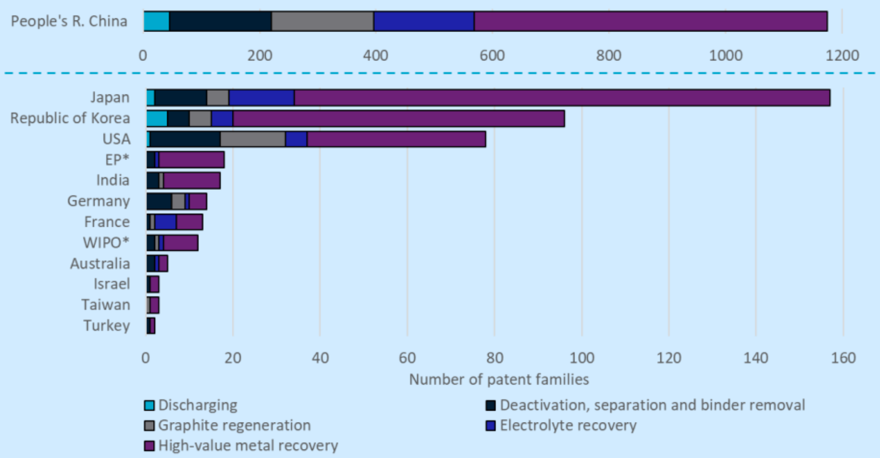
Figure 4: Taxonomy of battery recycling technologies.



Global R&D and commercialisation snapshot

Li-B recycling

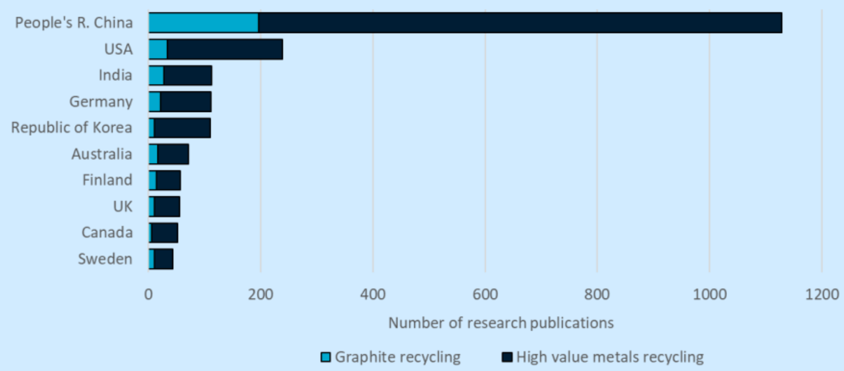
Figure 5: Patent output in battery recycling technologies from 2007 to 2022, by country.



*Applications filed under an entity other than a country.

People's R. China; People's Republic of China; WIPO, World Intellectual Property Organisation; EP, European Patent Office

Figure 6: Research publication output related to battery recycling, by country, over the 2007 – 2023 period.



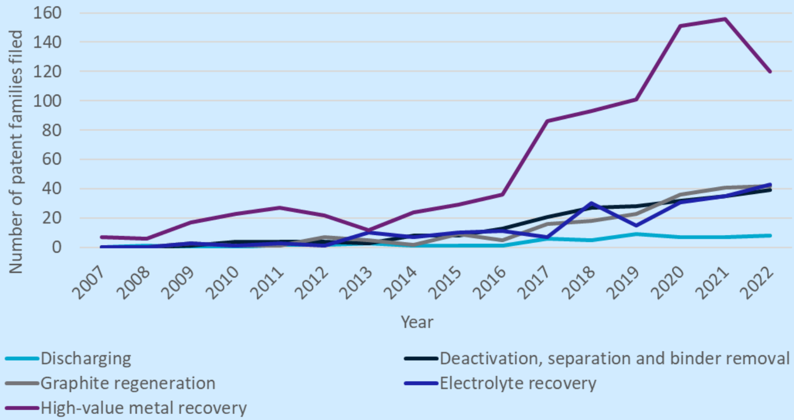
LIB, lithium-ion battery.

Figure 5 illustrates the global patent output for battery recycling technologies by country, between 2007 and 2022. China had the highest proportion across all domains, with the country's patent families accounting for 73.1% of global activity, followed by Japan's 9.8%, and the Republic of Korea's 6% of filings. Australia ranked 7th with patents across deactivation, separation and binder removal, electrolyte recovery, and high value metal recovery.

Figure 6 shows the research publication output related to battery recycling by country, for the 2007 – 2023 interval. Publications from China correspond to 45.6% of the total, followed by the USA (10%), and South Korea (4.9%). Australia ranked 6th with 2.6% of total publications.

The bibliometric and patent data presented in this report is subject to limitations and has an estimated accuracy of 70% or above. For a full description of the methodology and limitations refer to the main report *From minerals to materials: Assessment of Australia's critical mineral mid-stream processing capabilities*.

Figure 7: Patent output in battery recycling technologies from 2007 to 2022, by technology.



The recovery of high value metals was the area with the most patent activity, accounting for 56.7% of total patents in the battery recycling field. In contrast, patents related to the discharging process accounted for the lowest percentage of filed patents (3.3%). Electrolyte recovery (12.9%), graphite regeneration (13%) and dismantling and separation processes (14.1%) all attracted a similar volume of patents, following a similar upward trajectory after 2016 (Figure 7).

Table 1: Top 10 active organisations outside of China

By research publication output	By patent output
DOE, **United States**	JX Nippon Mining and Metals, **Japan**
IIT, **India**	Sumitomo metal Mining, **Japan**
Aalto University, **Finland**	SK Innovation, **Republic of Korea**
CNRS, **France**	Umicore, **Belgium**
Argonne National Laboratory, **USA**	Dow Eco System, **Japan**
Helmholtz Association, **Germany**	Mitsubishi Materials, **Japan**
CSIR, **India**	KIGAM, **Republic of Korea**
Chalmers University of Technology, **Sweden**	Attero Recycling PVT, **India**
University of California, **United States**	BASF, **Germany**
Sapienza University Rome, **Italy**	Cosmo Chemical, **Republic of South Korea**

The bibliometric and patent data presented in this report is subject to limitations and has an estimated accuracy of 70% or above. For a full description of the methodology and limitations refer to the main report *From minerals to materials: Assessment of Australia's critical mineral mid-stream processing capabilities*.

3.1 Discharging

Discharging is a method that involves removing the remaining charge within a collected battery, ensuring that spent batteries can be safely dismantled and separated. This reduces the risk of a thermal event caused by an uncontrolled release of stored electrical energy.[5] This is particularly important for large batteries or battery packs (e.g. electric vehicle batteries[6]), although this is not practiced consistently across industry. For smaller batteries (e.g. consumer electronics), recyclers opt to skip discharging and directly move to the next step of crushing and pyrolyzing the batteries, however the risks are still present.[7] However, the growing volumes of large batteries in waste streams will increase the importance of discharging.

Wet chemical discharging

Wet chemical discharging involves submerging the battery into a saltwater bath, such as a sodium chloride solution, to short circuit the battery.[8] Wet chemical discharging is widely utilised due to the simple operation and the low cost and accessibility of the salt solutions.[9] Sodium chloride is used due to the fast discharge rates achieved, however faces waste and safety issues related to chlorine and hydrogen gas emissions.[10] Further, electrolyte components can enter the solution, and materials can become contaminated. Wet chemical methods are commonly used on small-sized batteries.[11]

Electrical discharging

Electrical discharging uses an ohmic resistor or external circuits to transfer the remaining charge to external energy storage.[12] This approach is well suited for large-sized batteries, i.e. stationary energy storage systems or electric vehicle batteries.[13] This method has several advantages over wet chemical charging methods, such as safety and lower waste. However, is currently hard to scale due to different non-standardised battery designs, limiting process automation options.[14]

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[5] Yu et al. (2021) Pretreatment options for the recycling of spent lithium-ion batteries: A comprehensive review. Minerals Engineering 173, 107218.

[6] The most common battery cell used in EV's is the 18650 (10 watt hours, 3.6 volts). EV battery packs contain multiple battery cells.

[7] Windisch-Kern et al. (2022) Recycling chains for lithium-ion batteries: A critical examination of current challenges, opportunities and process dependencies. Waste Management.

[8] Zhao et al. (2021) Australian landscape for lithium-ion battery recycling and reuse in 2020 - current status, gap analysis and industry perspectives. CSIRO and FBI CRC.

[9] Zhao et al. (2021) Australian landscape for lithium-ion battery recycling and reuse in 2020 - current status, gap analysis and industry perspectives. CSIRO and FBI CRC.

[10] Wang et al. (2022) An effective and cleaner discharge method of spent lithium batteries. Journal of Energy Storage, 54, 105383.

[11] Zhao et al. (2021) Australian landscape for lithium-ion battery recycling and reuse in 2020 - current status, gap analysis and industry perspectives. CSIRO and FBI CRC.

[12] Windisch-Kern et al. (2022) Recycling chains for lithium-ion batteries: A critical examination of current challenges, opportunities and process dependencies. Waste Management.

[13] Windisch-Kern et al. (2022) Recycling chains for lithium-ion batteries: A critical examination of current challenges, opportunities and process dependencies. Waste Management.

[14] Zhao et al. (2021) Australian landscape for lithium-ion battery recycling and reuse in 2020 - current status, gap analysis and industry perspectives. CSIRO and FBI CRC.

Physical discharging

Physical discharging involves burying the battery in a solid conductive medium, such as copper powder, graphite powder, iron powder, or steel chips, to short-circuit the battery.[15] For example, physical discharging using flake graphite achieves a high discharge rate (comparable to wet chemical discharging with sodium chloride) while having low residual energy after discharge and zero pollution.[16] Technical challenges to scale up include ensuring thermal safety and storage methods for the powder.[17]

TECHNOLOGY STATE OF PLAY

Wet chemical discharging in sodium chloride, or other solutions, is commercially mature and is the most commonly deployed discharging method in industry.[18]

Electrical discharging is a mature technology that uses off the shelf equipment; however, industry adoption is at its early stages. Volkswagen's battery recycling pilot plant in Salzgitter, Germany opened in 2021. The batteries undergo electrical discharge/depletion, followed by processing through mechanical separation and hydrometallurgy.[19] Australian examples include a project at CSIRO developing an electrical discharging system that is compatible with batteries of all sizes and types. The energy extracted from the batteries can be fed into the grid or to the facility mains, reducing the energy usage and costs of the process.[20]

Physical discharging using conductive powders is currently at lab scale.[21]

An analysis of global patent filing activity shows that from 2007 to 2022, patents relating to discharging accounted for only 3% of patents that were filed in the battery recycling area. Trends show a slight increase in activity beginning after 2017. This indicates that discharging methods (particularly electrical and wet chemical discharge technologies) are highly mature with limited potential for new IP development, despite not being adopted as standard practice across industry and regulatory frameworks.

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[15] Yu et al. (2021) Pretreatment options for the recycling of spent lithium-ion batteries: A comprehensive review. Minerals Engineering 173, 107218; Gratz E, Sa Q, Apelian D, Wang Y (2014) A closed loop process for recycling spent lithium ion batteries. Journal of Power Sources 262, 255–262; Nan J, Han D, Zuo X (2005) Recovery of metal values from spent lithium-ion batteries with chemical deposition and solvent extraction. Journal of Power Sources, 152, 278-284.

[16] Wang et al. (2022) An effective and cleaner discharge method of spent lithium batteries. Journal of Energy Storage, 54, 105383.

[17] Yu et al. (2021) Pretreatment options for the recycling of spent lithium-ion batteries: A comprehensive review. Minerals Engineering 173, 107218.

[18] Kaya M (2022) State-of-the-art lithium-ion battery recycling technologies. Circular Economy 1(2), 100015.

[19] Volkswagen Group Components (2021) Battery recycling: Facts and figures about the pilot plant in Salzgitter. <https://www.volkswagen-newsroom.com/en/publications/more/battery-recycling-facts-and-figures-about-the-pilot-plant-in-salzgitter-605>

[20] Zhao et al. (2021) Australian landscape for lithium-ion battery recycling and reuse in 2020 - current status, gap analysis and industry perspectives. CSIRO and FBI CRC.

[21] Yao LP, Zeng Q, Qi T and Li J (2020) An environmentally friendly discharge technology to pretreat spent lithium-ion batteries. Journal of Cleaner Production 245, 118820. DOI: 10.1016/j.jclepro.2019.118820; Yu D, Huang Z, Makuza B, Guo X and Tian Q (2021) Pretreatment options for the recycling of spent lithium-ion batteries: A comprehensive review. Minerals Engineering 173, 107218. DOI: 10.1016/j.mineng.2021.107218.

The following table summarizes the key RD&D areas of focus in the discharging of used batteries:

Table 2. Global RD&D focus areas for the discharging of used batteries.

RD&D FOCUS AREAS	
General	• Increasing fundamental understanding of the electrochemical mechanisms behind the discharging process.

3.2 Dismantling and separation

Dismantling and separation break down batteries into small components and remove binder materials (usually polymer) for easier and more efficient extraction of metals and other valuable materials. This is typically done via a combination of mechanical, physical and chemical processes. Dismantling and separation sometimes involves a risk mitigation step (deactivation) to put the battery in a less reactive state, as an alternative to discharging.

The separation processes described in this section refer to the initial separation stages of battery components. Further hydro/pyrometallurgical process to separate the individual high value metals present in those components will be covered in Section 3.5. The separation and further processing of graphite and electrolyte components is an emerging area which will be covered in Sections 3.3 and 3.4.

This report will not cover initial manual steps of disassembly (e.g. packaging and hardware removal) that precede the mechanical, thermal and chemical processes.

3.2.1 Physical separation

Once battery packaging and hardware components have been disassembled and removed (often a manual process), the battery cell battery cells undergo a mechanical crushing process using equipment like crushers, ball mills, or shredders to separate components and materials from one another.

Mechanical processes may involve a deactivation step before or during crushing which temporarily puts the battery in a less reactive state. This is commonly achieved by subjecting the battery to cryogenic temperatures (using solid CO_2 or liquid nitrogen) or an inert environment (i.e., without moisture and oxygen) before or during to stabilise and reduce the crushing process risk of ignition. It should be noted that while deactivation reduces risk levels, it does not discharge the battery or entirely eliminate the risk. Cryogenic deactivation can also enhance the separation of binders from the electrodes, as low temperatures turn the binders from being elastic to being glassy and easy to break off.[22]

Sorting uses differences in size and density, as well as electrical and magnetic properties, to separate cell components.[23] Owing to its simplicity, sieving is the most common physical sorting method used in industry. Larger sieves are designed to collect the binders and electrolyte, whilst smaller sieves collect the cathode and anode active materials.[24]

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[22] Wang et al. (2019) Separation of the cathode materials from the Al foil in spent lithium-ion batteries by cryogenic grinding. Waste Management.

[23] Neumann et al. (2022) Recycling of lithium-ion batteries—current state of the art, circular economy, and next generation recycling. Advanced energy materials 12(17), 2102917.

[24] Kim et al. (2021) A comprehensive review on the pretreatment process in lithium-ion battery recycling. Journal of Cleaner Production 294, 126329.

Other physical sorting methods include electromagnetic, gravity and forth flotation techniques.[25] Metals such as copper, aluminium and iron from various part of the cells, modules, or packs are easily collected using these physical separation methods.

3.2.2 Thermal separation

Thermal separation is a simple and scalable process that utilises heat to decompose the binder materials, offering an alternative to both physical and chemical separation.

Thermal separation with oxygen present (i.e. incineration), can completely remove the carbon and polymer-based binders, enhancing the lithium and cobalt recovery rate.[26] Steam energy recovered from the process can be used for generators.[27] However, electrolytes and all plastic components are not recoverable via this process.[28]

Thermal separation without oxygen present (i.e., pyrolysis) decomposes organic compounds and binders into usable materials such as fuel or chemical feedstock and detaches aluminium foil from the cathode.[29] It also allows electrolyte recovery via condensation (see Section 3.4). Pyrolysis can also be assisted with microwaves, which helps improve efficiency.[30]

3.2.3 Chemical separation

Chemical processes (or dissolution) can also be used to remove binder materials and improve the separation of components. In this process, the shredded materials are submerged into a solvent. The solvent may be heated to temperatures of 30°C or higher to enhance separation efficiency, depending on the solvent. [31] Organic agents, such as N-methyl-2-pyrrolidone (NMP), work effectively to dissolve the binder and separate the cathode from the aluminium sheet.[32]

The dissolution method, however, does have various drawbacks, including the potential for high costs and the generation of toxic products, particularly when applied at a commercial scale.[33]

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[25] Kim et al. (2021) A comprehensive review on the pretreatment process in lithium-ion battery recycling. Journal of Cleaner Production 294, 126329.

[26] Makuza B, Tian Q, Guo X, Chattopadhyay K and Yu D (2021) Pyrometallurgical options for recycling spent lithium-ion batteries: A comprehensive review. Journal of Power Sources 491, 229622. DOI: 10.1016/j.jpowsour.2021.229622.

[27] Makuza B, Tian Q, Guo X, Chattopadhyay K and Yu D (2021) Pyrometallurgical options for recycling spent lithium-ion batteries: A comprehensive review. Journal of Power Sources 491, 229622. DOI: 10.1016/j.jpowsour.2021.229622.

[28] Makuza B, Tian Q, Guo X, Chattopadhyay K and Yu D (2021) Pyrometallurgical options for recycling spent lithium-ion batteries: A comprehensive review. Journal of Power Sources 491, 229622. DOI: 10.1016/j.jpowsour.2021.229622.

[29] Makuza B, Tian Q, Guo X, Chattopadhyay K and Yu D (2021) Pyrometallurgical options for recycling spent lithium-ion batteries: A comprehensive review. Journal of Power Sources 491, 229622. DOI: 10.1016/j.jpowsour.2021.229622.

[30] Diaz F, Wang Y, Moorthy T and Friedrich B (2018) Degradation Mechanism of Nickel-Cobalt-Aluminum (NCA) Cathode Material from Spent Lithium-Ion Batteries in Microwave-Assisted Pyrolysis. Metals 8(8), 565. DOI: 10.3390/met8080565.

[31] Zhou M, Liaw S, Sun Q, He R (2021) Pretreatment, Recycling, and Regeneration Strategies of Cathode Active Materials from Spent Lithium Ion Batteries. Journal of Material Sciences & Engineering, 10(8) 615. <https://www.hilarispublisher.com/open-access/pretreatment-recycling-and-regeneration-strategies-of-cathode-active-materials-from-spent-lithium-ion-batteries.pdf>

[32] Qadir et al. (2023) Formal and informal E-waste recycling methods for lithium-ion batteries: advantages and disadvantages. Global E-Waste Management Strategies and Future Implications. Elsevier.

[33] Zhou et al. (2021) Pretreatment, Recycling, and Regeneration Strategies of Cathode Active Materials from Spent Lithium Ion Batteries. Journal of Material Sciences & Engineering.

Some emerging methods focus on separating electrode materials from the attached current collector metal sheets, whilst simultaneously dissolving the electrolytes into a solution.[34] For discussion on how the electrolyte components are extracted and subsequently recovered (see Section 3.4).

TECHNOLOGY STATE OF PLAY

Physical crushing and separation is utilised by several major battery recycling companies at commercial scale.[35] For example, SungEel in the Republic of Korea and Accurec in Germany.[36]

Australian companies active in the battery recycling industry employ a variety of mechanical dismantling and separation techniques.

- Neometals, through their joint venture with Primobius (Germany), shred batteries and use a physical process to separate components like casings, metal foils and plastic from the electrode materials, prior to hydrometallurgical processing.[37] This has been purchased by Mercedes-Benz for installation at its Kuppenheim plant in Germany.[38]
- Similarly, Envirostream have commercialised a process that incorporates a blend of mechanical and hydraulic separation techniques, achieving a recovery rate of approximately 95% for battery materials.[39] This is then exported as black mass for further processing.

The use of deactivation methods prior to, or during, crushing is at pilot scale. For example, Cirba Solutions (US, formerly Retriev Technologies) is known to have patented and implemented cryogenic crushing in its recycling process. [40] Their approach involves cooling lithium metal batteries and large LIBs to approximately –200°C using liquid nitrogen.[41] This cryogenic method is reportedly used for all highly reactive batteries they process.[42] Inert environments using nitrogen or CO_2 gas is an approach chosen by many recyclers, such as Duesenfeld (Germany), and Recupyl (France).[43]

Thermal separation is also used at commercial scales internationally. For example, Accurec (Germany) uses a method which involves pyrolysis in a vacuum furnace at 250°C.[44] Conversely, Umicore (Belgium) uses a two-step process where the battery materials are first heated up to 300°C to evaporate the electrolyte. The

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[34] Kim et al. (2021) A comprehensive review on the pretreatment process in lithium-ion battery recycling. Journal of Cleaner Production 294, 126329; He et al. (2019) A green process for exfoliating electrode materials and simultaneously extracting electrolyte from spent lithium-ion batteries. Journal of Hazardous Materials.

[35] Sojka R, Pan Q, Billmann L. (2020) Comparative study of Li-ion battery recycling processes. ACCUREC Recycling GmbH.

[36] Sojka R, Pan Q, Billmann L. (2020) Comparative study of Li-ion battery recycling processes. ACCUREC Recycling GmbH.

[37] Primobius GmbH (n.d.) Recycling process. <https://www.primobius.com/en-au/technology-services/recycling-process>

[38] ASX Announcement (2024) Mercedes-Benz Refinery Purchase Order. <https://announcements.asx.com.au/asxpdf/20240110/pdf/05z9s9tqbsj2jh.pdf>

[39] Envirostream. Frequently asked Questions. <https://envirostream.com.au/frequently-asked-questions/#:~:text=Envirostream's%20battery%20recycling%20technology%2C%20which,Importantly%2C%20no%20incineration%20is%20involve>

[40] Sojka R, Pan Q, Billmann L. (2020) Comparative study of Li-ion battery recycling processes. ACCUREC Recycling GmbH; Bae H, Kim Y (2021) Technologies of lithium recycling from waste lithium ion batteries: a review. Materials advances 2(10), 3234-3250; Sheth R et al (2023) The Lithium-Ion Battery Recycling Process from a Circular Economy Perspective – A Review and Future Directions. Energies 16(7), 3228. <https://www.mdpi.com/1996-1073/16/7/3228>; McLaughlin W, Adams TS (1999) Li reclamation process. Patent number: US5888463A.

[41] Sonoc A, Jeswiet J, Soo VK (2015) Opportunities to Improve Recycling of Automotive Lithium Ion Batteries. Procedia CIRP 29, 752–757.

[42] Cirba Solutions (n.d.) Primary Lithium Processing. Material Processing. <https://www.cirbasolutions.com/primary-lithium-processing/>

[43] Sommerville et al. (2021). A qualitative assessment of lithium ion battery recycling processes. Resources, Conservation and Recycling, 165, 105219.

[44] Georgi-Maschler et al. (2012) Development of a recycling process for Li-ion batteries. Journal of power sources 207, 173-182.

temperature is then increased to 700ºC in a pyrolysis environment to decompose the organic binders into usable fuels.[45] Thermal separation methods are not currently utilised in Australia outside of lab scale research.

Chemical separation (or dissolution) is currently being used at commercial scale outside Australia but remain at lab scale in Australia. International examples include:

- Ascend Elements (US), which uses a patented process where the aluminium current collector is dissolved in sodium hydroxide after the initial shredding step. Drying, sieving and separation in a dense medium then allow the copper current collector to be retrieved. A sequence of hydrometallurgical steps on the residual mix removes plastics, carbon, iron, and the remaining aluminium and copper, leaving behind a solution of cobalt, nickel, and manganese for further processing.[46]
- In 2023, the company opened a facility in Georgia (US) capable of processing 30,000 metric tons of recyclable battery material.[47]

The patent filing analysis from 2007 to 2022, found global patent output related to the dismantling, and separation of LIBs has been increasing steadily over time, with an uptick in activity since 2016. China led the way with 77% of patent families, followed by the US at 7%, and Japan at 5%. Other active countries include Germany and the Republic of Korea. Australia has also been producing patents in this area. However, output is relatively low (less than 3% of global output). Australian organisations with patent activity in this area include NewSouth Innovations and Resource Conservation & Refining Corporation (subsidiary of Lithium Australia).

[45] Makuza et al. (2021) Pyrometallurgical options for recycling spent lithium-ion batteries: A comprehensive review. Journal of Power Sources 491, 229622.

[46] Gratz E, Sa Q, Apelian D, Wang Y (2014) A closed loop process for recycling spent lithium ion batteries. Journal of Power Sources 262, 255–262; Velázquez-Martínez O, Valio J, Santasalo-Aarnio A, Reuter M, Serna-Guerrero R (2019) A Critical Review of Lithium-Ion Battery Recycling Processes from a Circular Economy Perspective. Batteries 5(4):68; Ascend Elements (n.d.) Patented Hydro-to-Cathode® direct precursor synthesis process increases material performance and value. Innovation. <https://ascendelements.com/innovation/>

[47] Ascend Elements (2023) Ascend Elements Opens One of North America’s Largest Electric Vehicle Battery Recycling Facilities in Georgia. News & Insights. <https://ascendelements.com/ascend-elements-base-1-grand-opening/>

The following table summarizes the key RD&D areas of focus in the dismantling and separation of used batteries:

Table 3. Global RD&D focus areas for the dismantling and separation of used batteries.

RD&D FOCUS AREAS	
Physical separation	• Reducing the capital and equipment costs of cryogenic deactivation.[48] • Reducing and mitigating the contamination risks of the sieving method.[49]
Thermal separation	• Optimising the energy usage of thermal separation and electrifying thermal processes where possible to enable the integration of renewable energy.
Chemical separation	• Developing separation processes that replace toxic solvents, minimise hazardous by-products, and enable binder recovery.[50]
Supporting research domains	• Utilising advanced robotics, automation and artificial intelligence in the dismantling, sorting and separation steps to increase efficiency.[51]

3.3 Graphite separation and regeneration

Scarcity and global supply chain risks have spurred development in graphite separation and regeneration for use in battery anodes. To date, graphite separation and regeneration hasn't been the focus of existing LIBs recycling methods due to its lower value relative to other battery metals. [52] In current commercial processes the anodes, which contain battery grade graphite, are typically either incinerated for energy generation or disposed of in landfills.[53] More recently, however, research is being conducted to explore processes that regenerate graphite for use in battery anodes rather than discarding or downcycling the material for lower value uses.

In cases where is not economically or technically feasible to regenerate the graphite due to structural damage, research efforts are focused on conversion to high value functional materials (e.g., graphene and

[48] Ali H, Khan A, Pecht M (2022) Preprocessing of spent lithium-ion batteries for recycling: Need, methods, and trends. Renewable and Sustainable Energy Reviews 168, 112809.

[49] Kim et al. (2021) A comprehensive review on the pretreatment process in lithium-ion battery recycling. Journal of Cleaner Production, 294, 126329.

[50] Fu et al. (2021) Innovative recycling of organic binders from electric vehicle lithium-ion batteries by supercritical carbon dioxide extraction. Resources, Conservation and Recycling 172, 105666; Zhou et al. (2021) Pretreatment, Recycling, and Regeneration Strategies of Cathode Active Materials from Spent Lithium Ion Batteries. Journal of Material Sciences & Engineering 10, 8.

[51] Zhao et al. (2021) Australian landscape for lithium-ion battery recycling and reuse in 2020 - current status, gap analysis and industry perspectives. CSIRO and FBI CRC.

[52] Moradi B, Botte G (2016) Recycling of graphite anodes for the next generation of lithium-ion batteries. J Appl Electrochem 46, 123–148. <https://doi.org/10.1007/s10800-015-0914-0>

[53] Natarajan S, Divya L, Aravindan V (2022) Should we recycle the graphite from spent lithium-ion batteries? The untold story of graphite with the importance of recycling. Journal of Energy Chemistry, 71, 351-369.

composites) and low value utilisation (such as adsorbents for environmental remediation or reducing agents).[54] High value conversion and low value utilisation will not be covered in this chapter.

3.3.1 Froth flotation

Froth flotation is a commercial process used in mining that selectively separates materials by leveraging differences in their capacity to interact with water (surface hydrophobicity, or wettability).[55] Froth flotation is used after the crushing process to separate the hydrophobic graphite from the hydrophilic cathode materials. In this processes, mixed material is stirred in a flotation cell causing the graphite to rise to the surface, where it can then be collected for further processing.[56] Prior to flotation the battery undergoes chemical, thermal, or physical pre-treatment (see *3.2 Dismantling and separation*) to remove other materials (e.g., binders) and enhance the efficiency of the process.[57] Froth flotation methods have been observed to successfully recover graphite, with some studies showing over 97% recovery rates.[58]

3.3.2 Hydro- and pyro-metallurgical regeneration

A combination of hydrometallurgical and pyrometallurgical processes are being developed for regenerating graphite for battery applications.

Generally, hydrometallurgical steps are used to remove impurities from the graphite, for example leaching with sulphuric or hydrochloric acid.[59] Citric acid and deep eutectic solvents are also being considered as alternatives for leaching given their lower corrosivity as compared to mineral acids.[60] While citric acid and eutectic solvents are generally considered environmentally safe, eutectic solvents have had mixed results relating to their toxicity.[61]

Pyrometallurgical steps, such as smelting, calcination or sintering, are used to remove impurities and restore the crystalline structure of the graphite (graphitization), enhancing its performance as a battery

[54] Abdollahifar M (2022) Graphite Recycling from End-of-Life Lithium-Ion Batteries: Processes and Applications. Advanced Materials Technologies, 8(2), 2200368; Liu et al. (2022). Critical strategies for recycling process of graphite from spent lithium-ion batteries: A review. Science of the Total Environment, 816, 151621

[55] Vanderbruggen et al. (2021) A contribution to understanding the flotation behavior of lithium metal oxides and spheroidized graphite for lithium-ion battery recycling. Colloids and Surfaces A: Physicochemical and Engineering Aspects, 626 127111.

[56] Shin et al. (2020) Electrochemical performance of recycled cathode active materials using froth flotation-based separation process. Journal of The Electrochemical Society 167(2), 020504.

[57] Vanderbruggen et al. (2021) A contribution to understanding the flotation behavior of lithium metal oxides and spheroidized graphite for lithium-ion battery recycling. Colloids and Surfaces A: Physicochemical and Engineering Aspects, 626 127111.

[58] Vanderbruggen et al. (2021) A contribution to understanding the flotation behavior of lithium metal oxides and spheroidized graphite for lithium-ion battery recycling. Colloids and Surfaces A: Physicochemical and Engineering Aspects, 626 127111.

[59] Gao et al. (2020) Graphite recycling from the spent lithium-ion batteries by sulfuric acid curing–leaching combined with high-temperature calcination. ACS Sustainable Chemistry & Engineering 8(25), 9447-9455.

[60] Niu B, Xiao J, Xu Z. (2022) Advances and challenges in anode graphite recycling from spent lithium-ion batteries. Journal of Hazardous Materials, 439, 129678.

[61] Martínez M, Townley G, Martínez-Espinosa M (2022) Controversy on the toxic nature of Deep eutectic solvents and their potential contribution to environmental pollution. Heliyon.

anode.[62] Variations on these processes, such as microwave calcination, are being considered to overcome some of the energy demands of traditional pyrometallurgical processes.[63]

Hydrometallurgical and pyrometallurgical processes have shown the ability to achieve a high purity material that exhibit good electrochemical performance, with respects to both charge capacity and cycle life.[64] However, there are challenges in regenerating graphite which centre around removing impurities. Impurities in the graphite can alter the interaction between the anode and the electrolyte and affect the formation of a solid electrolyte interface (SEI), influencing long-term cycling stability and cell performance.[65] Developing processes that effectively deal with these challenges, whilst simultaneously minimising pollution and remaining cost-effective is essential.[66]

TECHNOLOGY STATE OF PLAY

The regeneration of graphite materials is currently at lab scale with plans to move to pilot scales. International examples include the Ascend Elements and Koura (US) joint 'Hydro-to-Anode' graphite recycling project. The technology has the potential to produce 99.9% pure graphite from end-of-life LIBs and battery manufacturing scrap.[67] Their patented process removes impurities via leaching while maintaining the dissolved recoverable metals, and reportedly can streamline the overall recycling process.[68]

Following successful testing results, Australian based Ecograf have progressed plans to build a modular recycling pilot plant to recover carbon battery anode materials using their hydrofluoric acid-free graphite purification technology. The process recycles anode material containing natural and synthetic graphite and can be reused in batteries when blended with EcoGraf's high purity Epanko (Tanzania) battery anode material.[69]

The patent analysis identified that 13% of patent families from 2007 to 2022 were related to the separation and regeneration of graphite from spent LIBs. This research area experienced strong IP activity in the last 3 years, indicating a growing interest in this domain and concern for circularity and resource scarcity. China filed 84% of patent families, followed by the US at 7%, and Japan and the Republic of Korea at 2%. Germany is also active in the area, albeit with smaller identified activity.

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[62] Gao et al. (2020) Graphite recycling from the spent lithium-ion batteries by sulfuric acid curing–leaching combined with high-temperature calcination. ACS Sustainable Chemistry & Engineering 8(25), 9447-9455.

[63] Fan et al. (2022) Regeneration of graphite anode from spent lithium-ion batteries via microwave calcination. Journal of Electroanalytical Chemistry 908, 116087.

[64] Liu et al. (2022). Critical strategies for recycling process of graphite from spent lithium-ion batteries: A review. Science of the Total Environment, 816, 151621; Gao et al. (2020) Graphite recycling from the spent lithium-ion batteries by sulfuric acid curing–leaching combined with high-temperature calcination. ACS Sustainable Chemistry & Engineering 8(25), 9447-9455.

[65] Fink et al. (2022) Influence of metallic contaminants on the electrochemical and thermal behavior of Li-ion electrodes. Journal of Power Sources 518, 230760; Berrueta et al. (2019) Lithium-ion batteries as distributed energy storage systems for microgrids. In Distributed Energy Resources in Microgrids (pp. 143-183). Academic Press.

[66] Liu et al. (2022). Critical strategies for recycling process of graphite from spent lithium-ion batteries: A review. Science of the Total Environment, 816, 151621.

[67] Ascend Elements (n.d.) Patented Hydro-to-Cathode® direct precursor synthesis process increases material performance and value. <https://ascendelements.com/innovation/>

[68] Ascend Elements (n.d.) Patented Hydro-to-Cathode® direct precursor synthesis process increases material performance and value. <https://ascendelements.com/innovation/>

[69] EcoGraf Limited (2023) SungEel Hitech Recycled Anode Material Performance. <https://www.ecograf.com.au/wp-content/uploads/2023/03/2529646.pdf>

The following table summarizes the key RD&D areas of focus in the separation and regeneration of graphite from used batteries:

Table 4. Global RD&D focus areas for the separation and regeneration of graphite from used batteries.

RD&D FOCUS AREAS	
Separation and regeneration of graphite	• Enhancing froth flotation efficiency by enhancing the pre-treatment step (separation and binder removal).[70] • Optimising hydrometallurgical processes to improve recovery efficiency, reduce wastewater and reagent consumption, lower equipment costs.[71] • Optimising the energy use of pyrometallurgical processes, and further development of emerging processes potential to reduce energy requirements (e.g. microwave irradiation).[72] • Mitigating the carbon loss that can occur in ultra-high temperature pyrometallurgical processes.[73]

3.4 Electrolyte recovery

The recovery of electrolytes is a growing area of interest in battery recycling, aiming to enhance material recovery and to enable an additional revenue stream from the recycling process, a key challenge given the low price for this material. Battery electrolyte typically comprises one or more organic solvents and one or more lithium salts. Electrolytes must be recovered at the upstream separation stages because the electrolyte salts will decompose under aggressive hydrometallurgical and pyrometallurgical processes used to recover other materials. The main techniques to achieve this include distillation, solvent extraction and supercritical CO_2 extraction.

Distillation is a process of evaporation and condensation to separate components leveraging the difference between their individual boiling points. Distillation can recover the solvent components of the electrolyte, however it requires managing toxic and corrosive gas pollution.[74] For example, for batteries undergoing pyrolytic thermal separation (described in Section 3.2.2), the lithium electrolyte salt lithium hexafluorophosphate ($LiPF_6$) decomposes into solid lithium fluoride (LiF) and toxic gases (i.e., hydrofluoric acid, phosphoric acid). LiF can be retrieved during the subsequent metallurgical processing of black mass.[75]

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[70] Niu B, Xiao J, Xu Z (2022) Advances and challenges in anode graphite recycling from spent lithium-ion batteries. Journal of Hazardous Materials 439, 129678.

[71] Liu et al. (2022) Critical strategies for recycling process of graphite from spent lithium-ion batteries: A review. Science of the Total Environment, 816, 151621.

[72] Niu B, Xiao J, Xu Z. (2022) Advances and challenges in anode graphite recycling from spent lithium-ion batteries. Journal of Hazardous Materials, 439, 129678.

[73] Niu B, Xiao J, Xu Z. (2022) Advances and challenges in anode graphite recycling from spent lithium-ion batteries. Journal of Hazardous Materials, 439, 129678.

[74] Niu et al. (2023) Recycling Hazardous and Valuable Electrolyte in Spent Lithium-Ion Batteries: Urgency, Progress, Challenge, and Viable Approach. Chemical Reviews 123(13), 8718–8735.

[75] Niu et al. (2023) Recycling Hazardous and Valuable Electrolyte in Spent Lithium-Ion Batteries: Urgency, Progress, Challenge, and Viable Approach. Chemical Reviews 123(13), 8718–8735.

Due to the application of heat, lithium salts will likely decompose and are typically not recoverable in their original state.

An alternative to distillation is solvent extraction followed by precipitation. Solvent extraction is a separation technique that uses the solubility differences to separate components of a mixture. Following the chemical separation process (see Section 3.2.3), solvent extraction can separate and extract the entire electrolyte from the LIB waste. The electrolyte components, such as the $LiPF_6$ can be separated in further downstream processing, via precipitation. Alternatively, the lithium can be recovered as lithium carbonate (Li_2CO_3).[76]

The pathway of precipitating $LiPF_6$ salt is being investigated due to the ability to recover both the solvent and salt components of the electrolyte. Further, the dissolving agent (typically organic carbonate) can be recycled via distillation for reuse at the end of the process.[77] However, when performed on shredded materials, the recovered electrolyte solvents and salts might be contaminated with impurities and require further purification.

Supercritical CO_2 extraction involves reacting shredded materials with CO_2 at specific temperatures and pressures in a closed-loop environment.[78] It is an emerging electrolyte recovery method with many advantages including high extraction efficiency, mild extraction conditions and low toxicity and pollution.[79] Currently, supercritical CO_2 only allows for electrolyte solvent recovery. However, by also adding other organic solvents the electrolyte (solvents and salts) can potentially be recovered in its almost original composition and the extraction duration can be reduced.[80] Low processing capacity and high investment costs for specialised equipment that can achieve and maintain constant temperatures and pressures are two challenges that remain in order to scale up this process.[81]

TECHNOLOGY STATE OF PLAY

Electrolyte recovery processes have been developed by several organisations however, there is limited information regarding the progress of demonstration on shredded battery materials.[82] Internationally, Accurec, Germany, has patented a process where electrolyte solvents are evaporated under a vacuum, separated from contaminants, and condensed for recovery.[83] This method leverages the pyrometallurgical step of the company's conventional recycling process, which heats disassembled batteries to 600°C to

[76] Niu et al. (2023) Recycling Hazardous and Valuable Electrolyte in Spent Lithium-Ion Batteries: Urgency, Progress, Challenge, and Viable Approach. Chemical Reviews 123(13), 8718–8735. DOI: 10.1021/acs.chemrev.3c00174.

[77] Niu et al. (2023) Recycling Hazardous and Valuable Electrolyte in Spent Lithium-Ion Batteries: Urgency, Progress, Challenge, and Viable Approach. Chemical Reviews 123(13), 8718–8735. DOI: 10.1021/acs.chemrev.3c00174.

[78] Niu et al. (2023) Recycling Hazardous and Valuable Electrolyte in Spent Lithium-Ion Batteries: Urgency, Progress, Challenge, and Viable Approach. Chemical Reviews 123(13), 8718–8735. DOI: 10.1021/acs.chemrev.3c00174.

[79] Niu et al. (2023) Recycling Hazardous and Valuable Electrolyte in Spent Lithium-Ion Batteries: Urgency, Progress, Challenge, and Viable Approach. Chemical Reviews 123(13), 8718–8735. DOI: 10.1021/acs.chemrev.3c00174.

[80] Grützke et al. (2015) Extraction of lithium-ion battery electrolytes with liquid and supercritical carbon dioxide and additional solvents. RSC Advances; Liu et al. (2014) Supercritical CO2 extraction of organic carbonate-based electrolytes of lithium-ion batteries. RSC Advances; Zhang et al. (2022) Organic Electrolytes Recycling From Spent Lithium-Ion Batteries. Global Challenges 6(12), 2200050.

[81] Niu et al. (2023) Recycling Hazardous and Valuable Electrolyte in Spent Lithium-Ion Batteries: Urgency, Progress, Challenge, and Viable Approach. Chemical Reviews 123(13), 8718–8735. DOI: 10.1021/acs.chemrev.3c00174.

[82] Sojka et al. (2020) Comparative study of Li-ion battery recycling processes. ACCUREC Recycling GmbH. <https://accurec.de/wp-content/uploads/2021/04/Accurec-Comparative-study.pdf>

[83] Sojka R, Melber A (2022) Method for decomposition of electrochemical storage devices and thermal treatment device. Patent number: EP3836290B1. <https://patents.google.com/patent/EP3836290B1/en?oq=EP3836290B1>

remove organic compounds.[84] OnTo Technology's (USA) supercritical CO2 electrolyte recovery technology is part of a patented direct recycling process which the company licenses and partners with recyclers to demonstrate (discussed more in Section 3.5).[85]

In Australia, CSIRO has developed and patented a simple, cost-effective process to recover electrolyte salt using solvent extraction and precipitation. The recovered products can be directly used to make new battery electrolytes.[86]

Globally, patent families related to electrolyte recovery have experienced significant increase since 2017. This reflects a growing trend in improved material recovery, going beyond the recovery of CAM metals. China accounted for 83% of patent families, followed by Japan with 7%, and France, the Republic of Korea, and the US with 2.4%.

The following table summarizes the key RD&D areas of focus in electrolyte recovery:

Table 5. Global RD&D focus areas for electrolyte recovery technologies.

RD&D FOCUS AREAS	
Electrolyte recovery	• Improving the extraction yield and purity of electrolytes in organic solvent extraction processes, whilst minimising solvent consumption.[87] • Developing strategies to manage toxic and corrosive gas emissions (particularly in the distillation methods).[88] • Optimising the use of organic solvents during supercritical CO_2 extraction to enable lithium salt recovery and reduce extraction time.[89]

3.5 High value metal recovery

The impetus to recover metals such as lithium, cobalt, and nickel from spent batteries is multifaceted. The concentration of these high value metals within batteries is higher than in natural ores.[90] Furthermore, recovering metals from spent batteries is a simpler process than extraction from primary resources.[91] Environmental concerns surrounding ecotoxicity, and supply concerns such as resource scarcity have also

[84] Sojka et al. (2020) Comparative study of Li-ion battery recycling processes. ACCUREC Recycling GmbH. <https://accurec.de/wp-content/uploads/2021/04/Accurec-Comparative-study.pdf>

[85] Sloop S, Crandon L, Allen M, Koetje K, Reed L, Gaines L, Sirisaksoontorn W and Lerner M (2020) A direct recycling case study from a lithium-ion battery recall. Sustainable Materials and Technologies 25, e00152. DOI: 10.1016/j.susmat.2020.e00152.

[86] Zhao et al. (2021) Australian landscape for lithium-ion battery recycling and reuse in 2020 - current status, gap analysis and industry perspectives. CSIRO and FBI CRC.

[87] Zhang et al. (2022) Organic Electrolytes Recycling From Spent Lithium-Ion Batteries. Global Challenges 6(12), 2200050.

[88] Niu et al. (2023) Recycling Hazardous and Valuable Electrolyte in Spent Lithium-Ion Batteries: Urgency, Progress, Challenge, and Viable Approach. Chemical Reviews 123(13), 8718–8735.

[89] Grützke et al. (2015) Extraction of lithium-ion battery electrolytes with liquid and supercritical carbon dioxide and additional solvents. RSC Advances.

[90] Makuza et al. (2021) Pyrometallurgical options for recycling spent lithium-ion batteries: A comprehensive review. Journal of Power Sources 491, 229622.

[91] Xiao et al. (2020) Challenges to Future Development of Spent Lithium Ion Batteries Recovery from Environmental and Technological Perspectives. Environmental Science & Technology 54(1), 9–25.

driven research in this area.[92] The recovery of high value metals provides the revenue stream required to support battery recycling operations. This is particularly attractive for battery chemistries such as NMC (nickel manganese cobalt oxide), which contain several high value metals (i.e. nickel and cobalt), however this will become commercially more challenging as manufacturers reduce the amount of high-cost metals in cathodes. Battery chemistries with fewer high value metals, such as LFP (lithium iron phosphate, where lithium is the main metal of interest), provide a smaller revenue stream.[93]

For all chemistries, RD&D efforts are focused developing commercially attractive recycling options. Pyrometallurgy and hydrometallurgy methods, or a combination of the processes, have been at the centre of efforts aimed at recovering high value metals.

3.5.1 Pyrometallurgical and hydrometallurgical extraction

Pyrometallurgy

Pyrometallurgical extraction entails the high temperature treatment of black mass to form metal oxides and compounds. Pyrometallurgical techniques can be applied on the most common electric vehicle (EV) battery chemistries (NMC, NCA – nickel cobalt aluminium oxide – and LFP), and is able to recover copper, cobalt, nickel and iron.[94] However, it may be less optimal for LFP batteries given this chemistry does not contain nickel and cobalt.

Smelting is the conventional pyrometallurgical extraction method due to its simplicity. The black mass is heated at temperatures above its melting point to produce a molten metal alloy. In the case of NMC batteries, this produces an alloy containing copper, cobalt, nickel and iron, and a slag of lithium, aluminium, manganese and other metals.[95] The lithium present in the slag is not further extracted and is currently wasted.[96] However, in recent years, there have been some research attempts to extract it from the slag via chloride or sulphate salt roasting, particularly with respect to recovering the lithium content of LFP batteries.[97]

An alternative pyrometallurgical extraction method is reduction roasting, which requires lower temperatures than smelting. Reduction roasting involves roasting the pre-treated CAM with a reducing agent, which could be a carbon source like charcoal or coke, a chlorination, sulfation or nitration salt, or oxygen, to form a lithium compound and a metal oxide. The product mixture is then leached with water to precipitate out the insoluble metal oxide and form an aqueous solution containing lithium, which is then evaporated to obtain the lithium compound.[98]

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[92] Makuza et al. (2021) Pyrometallurgical options for recycling spent lithium-ion batteries: A comprehensive review. Journal of Power Sources 491, 229622.

[93] Pesaran A et al (2023) Electric vehicle lithium-ion battery life cycle management. NREL. <https://www.nrel.gov/docs/fy23osti/84520.pdf>

[94] Dobó Z, Dinh T, Kulcsár T (2023) A review on recycling of spent lithium-ion batteries. Energy Reports, 9, 6362-6395.

[95] Makuza et al. (2021) Pyrometallurgical options for recycling spent lithium-ion batteries: A comprehensive review. Journal of Power Sources 491, 229622.

[96] Makuza et al. (2021) Pyrometallurgical options for recycling spent lithium-ion batteries: A comprehensive review. Journal of Power Sources 491, 229622.

[97] Li et al. (2019) Aqueous leaching of lithium from simulated pyrometallurgical slag by sodium sulfate roasting. RSC advances 9(41), 23908-23915; Dang et al. (2018) Recycled lithium from simulated pyrometallurgical slag by chlorination roasting. ACS Sustainable Chemistry & Engineering 6(10), 13160-13167.

[98] Bae H, Kim Y (2021) Technologies of lithium recycling from waste lithium-ion batteries: a review. Materials Advances; Makuza et al. (2021) Pyrometallurgical options for recycling spent lithium-ion batteries: A comprehensive review. Journal of Power Sources.

A variation of reduction roasting uses microwaves to improve extraction efficiency and reduce energy intensity, since carbon particles can effectively absorb microwave energy and raise temperature.[99]

Hydrometallurgy

Hydrometallurgical extraction methods use various solvents to extract metals from black mass and separate different CAM metals from each other. The most common EV battery types (NMC, NCA and LFP) can all be recycled via hydrometallurgical routes. Its ability to recover high purity materials and to recover lithium means it may be more suitable than pyrometallurgy for chemistries like LFP.[100]

Various hydrometallurgical-based extraction processes have been developed and commercialised globally. The energy intensity of hydrometallurgy can depend on the battery chemistry, and the number of purification steps undertaken.[101] Hydrometallurgy is being pursued due to its the high recovery rates, and ability to recovery value from low grade feedstock.[102]

To extract lithium, the pre-treated CAM is first leached with an acid or alkali to form a solution containing different metal ions. There are various separation and extraction methods used to extract pure metal fractions from this solution, including solvent extraction and electrodeposition or precipitation.[103] These processes are able to filter out the non-lithium ions, utilising the different size, solubility and reaction to pH and temperature that different metal ions have. The final solution only contains lithium ions, and a carbonated salt is added to it to obtain lithium carbonate.[104]

Despite its maturity, hydrometallurgical extraction is based on a lengthy solvent extraction process requiring multiple circuits and large volumes of reagents and generating large volumes of contaminated wastewater. Additional circuits and reagents are required if further purification steps are undertaken.

TECHNOLOGY STATE OF PLAY

Pyrometallurgical recycling is a mature battery recycling technology already in use today.[105]

- Umicore (Belgium) and Inmetco (US), who both recycle about 6,000 to 7,000 tonnes of batteries per year, use smelting technologies.[106] Umicore's smelting technology converts end-of-life battery materials into a metal alloy that includes cobalt, nickel, lithium, and copper. This alloy undergoes additional refinement in a hydrometallurgical process to recover the individual metals.[107]

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[99] Makuza et al. (2021) Pyrometallurgical options for recycling spent lithium-ion batteries: A comprehensive review. Journal of Power Sources.

[100] Vasconcelos et al. (2023) Circular Recycling Strategies for LFP Batteries: A Review Focusing on Hydrometallurgy Sustainable Processing. Metals, 13(3), 543.

[101] Stakeholder consultations.

[102] Zhao et al. (2021) Australian landscape for lithium-ion battery recycling and reuse in 2020 - current status, gap analysis and industry perspectives. CSIRO and FBI CRC.

[103] Zhao et al. (2021) Australian landscape for lithium-ion battery recycling and reuse in 2020 - current status, gap analysis and industry perspectives. CSIRO and FBI CRC.

[104] Bae H, Kim Y (2021) Technologies of lithium recycling from waste lithium-ion batteries: a review. Materials Advances 2(10), 3234-3250.

[105] Sommerville et al. (2021) A qualitative assessment of lithium-ion battery recycling processes. Resources, Conservation and Recycling 165, 105219.

[106] Dobó Z, Dinh T, Kulcsár T (2023) A review on recycling of spent lithium-ion batteries. Energy Reports, 9, 6362-6395.

[107] Umicore (n.d.) Our recycling process. <https://brs.umicore.com/en/recycling/>; Makuza et al. (2021) Pyrometallurgical options for recycling spent lithium-ion batteries: A comprehensive review. Journal of Power Sources.

- Reduction roasting has yet to be commercially applied in industry. In 2015 the EcoBatRec project, co-developed by RWTH Aachen University and Accurec (Germany), proposed carbothermic reduction to recover lithium from spent LIBs. The reduction was followed by vacuum evaporation to recover lithium metal. Alternatively, evaporation in a nitrogen environment with subsequent oxidation retrieved lithium oxide.[108]

Hydrometallurgical recycling is being deployed at commercial scales globally. Hydrometallurgical pathways are used in plants across Canada, China, France, the United States, and South Korea. Plants that combine pyro and hydrometallurgical methods have also been established in Belgium, Finland, Germany, Japan and Switzerland.[109] It is reported that more than 70 percent of the major recyclers in China use hydrometallurgical techniques as their primary process.[110] In 2023, the US commissioned its first commercial scale hydrometallurgical plant, Li-Cycle, the capacity to process up to 35,000 tonnes of materials per year. The company is also looking to expand its operational network to Europe.

Australian companies are in the process of demonstrating hydrometallurgical battery recycling plants.

- Neometals (Australia) utilises multiple solvents to extract valuable metals from used consumer electronic batteries (e.g., lithium cobalt oxide, LCO) and EV and energy storage system batteries (e.g., NMC). The company is demonstrating its technology in Germany through a joint venture with the German SMS group.[111]
- Green Li-ion (Singapore), has based its R&D operations at Deakin University, has developed a hydrometallurgical process capable of producing precursor cathode active material (pCAM) for NCM batteries, graphite, and lithium carbonate directly from black mass. The company commercialises its equipment and material recovery processes to companies around the world instead of providing direct LIB recycling services.

An analysis of global patents from 2007 to 2022 showed that over half of all battery recycling patents are related to pyrometallurgical and hydrometallurgical recovery of high value metals. This reflects the strong interest in these metals due to their attractive price point and importance to the economics of recycling operations. There has been a significant increase in patent activity for the area since 2017. China accounted for 67% of patent families, followed by the Japan with 14% and the Republic of Korea with 8%. Other countries with significant activity include the US and India.

Global IP activity for the direct recovery of CAM and bio-hydrometallurgical processes were not included in this report due to the emergence of these technologies and the limited number of patents being published globally.

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[108] Träger T, Friedrich B, Weyhe R (2015) Recovery concept of value metals from automotive lithium-ion batteries. Chemie ingenieur technik 87(11), 1550-1557.i

[109] Baum ZJ, Bird RE, Yu X, Ma J (2022) Lithium-Ion Battery Recycling–Overview of Techniques and Trends. ACS Energy Letters 7(2), 712–719.

[110] Research and Markets (2019) Growth opportunities in the circular economy for global electric vehicle battery reuse (second-life) and recycling market, forecast to 2025. <https://www.researchandmarkets.com/reports/4803957/growth-opportunities-in-the-circular-economy-for>

[111] Neometals Ltd (n.d.) Battery Recycling. <https://www.neometals.com.au/business-units/core-divisions/lib/>

The following table summarizes the key RD&D areas of focus in pyrometallurgical and hydrometallurgical extraction methods:

Table 6. Global RD&D focus areas for pyrometallurgical extraction methods.

RD&D FOCUS AREAS	
Pyrometallurgical extraction	• Overcoming the challenges associated with different reagents in reduction roasting (e.g., low lithium recovery rate when roasting with carbon, equipment corrosion and toxic gas emissions when roasting with a sulphate or chloride salt).[112] • Optimising the extraction and cost efficiency of metal recovery (including lithium) from smelted slag.[113] • Developing cost-effective solutions to treat toxic gas emissions (e.g., dioxins).[114]
Hydrometallurgical extraction	• Exploring alternative reagents (e.g., organic acids, ionic liquids, deep eutectic systems) to improve extraction efficiency and minimise reagent consumption and waste production.[115] • Designing a close-looped system to capture, treat and recycle the used solvents to reduce reagent cost and waste.[116] • Designing and optimising the operational process at scale, which requires precise control of reaction parameters.[117]

3.5.2 Electrochemical separation

The complexity and reagent intensity of conventional hydrometallurgical separation pathways have driven interest in electrochemical methods (e.g., electrodeposition) as an alternative for the recovery of high value metals from CAM leaching solutions.

Electrochemical separation selectively reduces ions of the metal of interest using an electric current, bringing it out of solution. Besides avoiding large reagent volumes and featuring a simpler process,

[112] Cornelio A, Zanoletti A and Bontempi E (2024) Recent Progress in Pyrometallurgy for the Recovery of Spent Lithium-Ion Batteries: A Review of State-of-the-Art Developments. Current Opinion in Green and Sustainable Chemistry 100881. DOI: 10.1016/j.cogsc.2024.100881.

[113] Latini D, Vaccari M, Lagnoni M, Orefice M, Mathieux F, Huisman J, Tognotti L and Bertei A (2022) A comprehensive review and classification of unit operations with assessment of outputs quality in lithium-ion battery recycling. Journal of Power Sources 546, 231979. DOI: 10.1016/j.jpowsour.2022.231979; Fan Y, Li H, Lu C, Chen S, Yao Y, He H, Ma S, Peng Z and Shao K (2023) A novel method for recovering valuable metals from spent lithium-ion batteries inspired by the mineral characteristics of natural spodumene. Journal of Cleaner Production 417, 138043. DOI: 10.1016/j.jclepro.2023.138043.

[114] Makuza et al. (2021) Pyrometallurgical options for recycling spent lithium-ion batteries: A comprehensive review. Journal of Power Sources.

[115] Saleem U, Joshi B and Bandyopadhyay S (2023) Hydrometallurgical Routes to Close the Loop of Electric Vehicle (EV) Lithium-Ion Batteries (LIBs) Supply chain: A Review. Journal of Sustainable Metallurgy 9(3), 950–971. DOI: 10.1007/s40831-023-00718-w; Zanoletti A, Carena E, Ferrara C and Bontempi E (2024) A Review of Lithium-Ion Battery Recycling: Technologies, Sustainability, and Open Issues. Batteries 10(1), 38. DOI: 10.3390/batteries10010038.

[116] Baum et al. (2022) Lithium-ion battery recycling– overview of techniques and trends. ACS Energy Letter 7 (2), 712–719.

[117] Bae H, Kim Y (2021) Technologies of lithium recycling from waste lithium-ion batteries: a review. Materials Advances 2(10), 3234-3250.

electrochemical separation can be powered by renewable energy sources, enhancing the sustainability of the process at large scale.[118] Challenges to overcome include improving the selectivity of the process.

The following table summarizes the key RD&D areas of focus in electrochemical separation methods:

Table 7. Global RD&D focus areas for electrochemical separation methods.

RD&D FOCUS AREAS	
Electrochemical separation	• Improving the electrode-electrolyte interface, designing electrochemical cells, and addressing realistic solution conditions to enhance the separation of difficult-to-deposit metals.[119] • Explore solvents (e.g., deep eutectic solvents) that enhance the selectivity of electrochemical separation and support CAM leaching integration.[120] • Develop strategies to reduce energy consumption and utilise side reactions for valuable by-product generation during electrochemical separation.[121]

TECHNOLOGY STATE OF PLAY

The electrochemical separation of multiple high value metals beyond lithium is being tested at pilot scale. Aqua Metals (USA) developed an electrochemical separation method called Li AquaRefining™, which reportedly can recover lithium hydroxide as well as copper, nickel, cobalt and manganese metal. In 2023, the company established a pilot facility with the intent of recycling 75 tonnes per annuum of battery materials.[122] Evonic (Germany) and NEU Battery Materials (Singapore) are exploring membrane electrolysis at pilot scale to separate lithium from leachate solutions and produce lithium hydroxide.[123]

While electrochemical separation is currently not investigated or utilised for LIB recycling in Australia, there are relevant capabilities from cross-cutting areas that can potentially be leveraged. Neometals (Australia) has developed and patented ELi™, an electrolysis process to extract lithium compounds from brines. Neometals is piloting the process through a joint venture with Mineral Resources (Australia) called Reed

[118] Prabaharan et al. (2017) Electrochemical process for electrode material of spent lithium ion batteries. Waste Management 68, 527–533; Kim et al. (2021) Selective cobalt and nickel electrodeposition for lithium-ion battery recycling through integrated electrolyte and interface control. Nature Communications 12(1), 6554; Kim et al. (2021) Electrochemical approaches for selective recovery of critical elements in hydrometallurgical processes of complex feedstocks. iScience 24(5), 102374.

[119] Kim et al. (2021) Electrochemical approaches for selective recovery of critical elements in hydrometallurgical processes of complex feedstocks. iScience 24(5), 102374.

[120] Kim et al. (2021) Electrochemical approaches for selective recovery of critical elements in hydrometallurgical processes of complex feedstocks. iScience 24(5), 102374.

[121] Li et al. (2023) Electrochemical methods contribute to the recycling and regeneration path of lithium-ion batteries. Energy Storage Materials 55, 606–630.

[122] AquaMetals (2024) Lithium Pilot Recycling Hub. Clean Recycling Innovator. <https://aquametals.com/pilot-recycling-hub/>

[123] NEU Battery Materials (2023) Redox Targeting Electrochemical extraction. Technology. <https://www.neumaterials.com/technology>; Ng G (2023) From waste to resource: NEU Battery Materials' electrochemical separation solution for lithium ion batteries. KrAsia. <https://kr-asia.com/from-waste-to-resource-neu-battery-materials-electrochemical-separation-solution-for-lithium-ion-batteries>; Evonik Industries (2022) Lithium from electric vehicle batteries: Moving towards better recycling. RD&I Press Release. <https://corporate.evonik.com/en/media/press-releases/corporate/lithium-from-electric-vehicle-batteries-moving-towards-better-recycling-177408.html>.

Advanced Materials.[124] Various membrane electrolysis processes and applications are also investigated domestically, such as to produce lithium hydroxide from lithium sulphate[125] and to recover ammonia from wastewater.[126]

3.5.3 Direct recovery of CAM

Direct recovery of CAM is an emerging approach that focuses on regenerating the cathode to restore its electrochemical performance.[127] This is achieved by recovering the initial composition and crystal structure of used CAM, rather than extracting the metals out of them. The pre-treated CAM is first mixed with additional lithium salts to make up for the lost lithium content before it is heated at high temperature in the presence of oxygen. Conductive agents like carbon or polymers can then be optionally added to the restored CAM to improve electrical conductivity.[128]

Direct recycling is being investigated as an alternative to pyro/hydrometallurgical processes due to its potential to close the material loop in cathode manufacturing, simple operation, lower levels of secondary waste and hazardous emissions.[129] However, the diverse and changing nature of LIBs chemical compositions may introduce operational, technical and economic challenges that inhibit the scaling of direct recycling processes.[130] For instance, sorting and separation procedures would be required to achieve a homogenous feedstock of battery chemistries, adding towards to labour intensity and cost of operations.[131] Stakeholder consultations suggest that this method may best be deployed alongside a battery manufacturing plant to process CAM manufacturing waste from a single source.

TECHNOLOGY STATE OF PLAY

The direct recovery of CAM is emerging with a limited number of global projects having reached pilot scale.

- Cathode-healing® is a patented direct recovery process invented by OnTo Technologies (US). The main reaction is conducted inside an autoclave with a subsequent thermal treatment to form renewed CAMs. In 2021, the company partnered with Johnson Matthey (UK) to demonstrate the technology at a pilot scale facility with a processing capacity of 1,000 tonnes per year.[132]

[124] Neometals (n.d.) Lithium chemicals. <https://www.neometals.com.au/en/business-units/core-divisions/lithium-chemicals/>.

[125] Chen X, Ruan X, Kentish SE, Li G (Kevin), Xu T and Chen GQ (2021) Production of lithium hydroxide by electrodialysis with bipolar membranes. Separation and Purification Technology 274, 119026. DOI: 10.1016/j.seppur.2021.119026.

[126] CSIRO (2024) Exploring the potential for ammonia recovery from wastewater with bipolar membrane electrodialysis. <https://research.csiro.au/hydrogenfsp/ammonia-from-wastewater/>.

[127] Ji et al. (2021) Direct recycling technologies of cathode in spent lithium-ion batteries. Clean Technol Recycl 1: 124–151.

[128] Ji et al. (2023) Direct regeneration of degraded lithium-ion battery cathodes with a multifunctional organic lithium salt. Nature Communications.

[129] Dobó Z, Dinh T, Kulcsár T (2023) A review on recycling of spent lithium-ion batteries. Energy Reports, 9, 6362-6395.; Ji et al. (2021). Direct recycling technologies of cathode in spent lithium-ion batteries. Clean Technol Recycl 1: 124–151.

[130] Pesaran et al. (2023) Electric Vehicle Lithium-Ion Battery Life Cycle Management. National Renewable Energy Laboratory. NREL/TP-5700-84520. <https://www.nrel.gov/docs/fy23osti/84520.pdf>

[131] Dobó et al. (2023) A review on recycling of spent lithium-ion batteries. Energy Reports, 9, 6362-6395.

[132] Johnson Matthey (2021) Johnson Matthey to improve the sustainability of Li-ion battery manufacturing with partners OnTo Technology and the UK Battery Industrialisation Centre. Johnson Matthey, UK; Sloop E (2021) Deactivation and Cathode-Healing™ to support the Low-Cost US Supply Chain. OnTo Technology LLC.

- A leading institution in this space is the ReCell Centre, which is a collaboration between six US national laboratories and universities. In 2022, the Centre led 12 projects related to the direct recovery process of CAM, covering from establishing best-practice and energy efficient processes to overcoming technical challenges (e.g., recovery efficiency and purity of outputs).[133]

The following table summarizes the key RD&D areas of focus in the direct CAM recovery methods:

Table 8. Global RD&D focus areas for direct CAM recovery methods.

RD&D FOCUS AREAS	
Direct CAM recovery	• Improving the flexibility of the process at scale, especially for dealing with more complex feedstocks (e.g. mixed CAM materials and CAM with a higher degradation due to high battery usage).[134] • Preserving CAM structure and reliably achieving high purity levels matching the requirements of new batteries.[135] • Optimising the initial sorting and component separation of batteries with different cathode chemistries, to enable single-material processing at scale.[136]

3.5.4 Bio-hydrometallurgical extraction

Bio-hydrometallurgical extraction uses the biological activities and byproducts of microorganisms to separate metals from used battery materials. Various microorganisms can produce low-toxicity acids that can dissolve metals into a solution or form metal-organic complexes, enabling subsequent metal precipitation.[137] Compared to the conventional pyrometallurgical or hydrometallurgical processes, bio-hydrometallurgical processes may offer advantages in terms of capital costs, energy consumption and environmental impact.[138]

Challenges to scaling up bio-hydrometallurgical techniques include leaching efficiency, long treatment times, large scale cultivation of microorganisms, and resistance to more aggressive conditions such as acidity and temperature levels.[139]

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133 ReCell Center (2022) The ReCell Center For Advanced Battery Recycling – FY22 Q4 Report. ReCell Center.

134 Harper et al. (2019) Recycling lithium-ion batteries from electric vehicles. Nature 575(7781), 75-86.

135 Bai et al. (2020) Energy and environmental aspects in recycling lithium-ion batteries: Concept of Battery Identity Global Passport. Materials Today 41, 304–315; Chen et al. (2019) Recycling End-of-Life Electric Vehicle Lithium-Ion Batteries Joule 3(11), 2622–2646,

136 Dobó Z, Dinh T, Kulcsár T (2023) A review on recycling of spent lithium-ion batteries. Energy Reports, 9, 6362-6395.

137 Wu et al. (2019) Mechanism underlying the bioleaching process of LiCoO2 by sulfur-oxidizing and iron-oxidizing bacteria. Journal of Bioscience and Bioengineering; Bahaloo-Horeh N, Mousavi SM (2017) Enhanced recovery of valuable metals from spent lithium-ion batteries through optimization of organic acids produced by Aspergillus niger. Waste Management.

138 Bahaloo-Horeh N, Mousavi SM (2017) Enhanced recovery of valuable metals from spent lithium-ion batteries through optimization of organic acids produced by Aspergillus niger. Waste Management; Bahaloo-Horeh N, Vakilchap F, Mousavi M (2019) Bio-hydrometallurgical methods for recycling spent lithium-ion batteries. Recycling of spent lithium-ion batteries: Processing methods and environmental impacts, 161-197.

139 Zheng et al. (2018) A Mini-Review on Metal Recycling from Spent Lithium Ion Batteries. Engineering 4(3), 361–370; Moazzam et al. (2021) Lithium bioleaching: An emerging approach for the recovery of Li from spent lithium-ion batteries. Chemosphere 277, 130196.

TECHNOLOGY STATE OF PLAY

Bio-hydrometallurgical recycling of LIB waste is an emerging pathway with relatively low technology maturity globally. Most laboratory-based research up till now has focused on the leaching technique, the type of microorganism used, the reaction conditions and the types of metal recovered.[140]

In Australia, CSIRO researchers have applied bio-hydrometallurgy to extract lithium, cobalt, nickel, manganese and copper from used laptop batteries at lab scale.[141] Capabilities across other sectors and applications can potentially be leveraged and applied to the battery recycling space. This includes bioleaching to extract high value metals e-waste and low-grade or complex ores.[142] Industry examples include Anax (Western Australia) developing IP and successfully testing bioleaching for potential commercial-scale extraction of copper from ores,[143] and Mint Innovation developing biorefineries for e-waste.[144]

The following table summarises the key RD&D areas of focus in the bio-hydrometallurgical extraction methods:

Table 9. Global RD&D focus areas for bio-hydrometallurgical extraction methods.

RD&D FOCUS AREAS	
Bio-hydrometallurgical extraction	• Increasing the knowledge about the interactions and metabolic mechanisms of the microorganisms.[145] • Improving the factors important for large-scale application, such as reaction efficiency (including the use of catalysts) and microorganism cultivation.[146] • Developing hybrid biological and chemical leaching techniques to improve extraction effectiveness.[147] • Developing microorganism strains to improve their tolerance for heavy metals, high temperatures and pH.[148]

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140 Bahaloo-Horeh N, Vakilchap F, Mousavi M (2019) Bio-hydrometallurgical methods for recycling spent lithium-ion batteries. Recycling of spent lithium-ion batteries: Processing methods and environmental impacts, 161-197.

141 Boxall NJ, Cheng KY, Bruckard W and Kaksonen AH (2018) Application of indirect non-contact bioleaching for extracting metals from waste lithium-ion batteries. Journal of Hazardous Materials 360, 504–511. DOI: 10.1016/j.jhazmat.2018.08.024.

142 CSIRO (2022) Microbes to mine metals from e-waste? <https://www.csiro.au/en/news/All/Articles/2021/December/Biomining>

143 Anax Metals (2023) Bioleaching success to boost Whim Creek metal production. <https://anaxmetals.com.au/wp-content/uploads/2023/06/2566518.pdf>; Reuters (2022) Miners turn to bacteria and other new ways to leach copper from waste rock. <https://www.reuters.com/markets/us/miners-turn-bacteria-other-new-ways-leach-copper-waste-rock-2022-05-11/>.

144 Mint (2024) A way to process e-waste that's better for business <https://www.mint.bio/solutions/biorefineries>

145 Moazzam et al. (2021) Lithium bioleaching: An emerging approach for the recovery of Li from spent lithium-ion batteries. Chemosphere 277, 130196.

146 Moazzam et al. (2021) Lithium bioleaching: An emerging approach for the recovery of Li from spent lithium-ion batteries. Chemosphere 277, 130196.

147 Moazzam et al. (2021) Lithium bioleaching: An emerging approach for the recovery of Li from spent lithium-ion batteries. Chemosphere 277, 130196.

148 Moazzam et al. (2021) Lithium bioleaching: An emerging approach for the recovery of Li from spent lithium-ion batteries. Chemosphere 277, 130196.

3.6 Implications for Australia

Australia's battery recycling industry is relatively small, with the majority of commercial recyclers focusing on collecting, crushing and separation to produce black mass, and exporting for further processing. A 2021 CSIRO report estimated Australia's LIB waste stream will reach approximately 136,000 tonnes of waste by 2036, with an associated recoverable value of $3 billion.[149] A more recent assessment by UTS and the Battery Council projects 360,000 tonnes by 2040 and 1.6 million tonnes by 2050.[150] Poor collection rates, recycling rates (only 10% in 2021), excessive landfilling of batteries and offshore recycling are all major factors that contribute to this economic loss.[151]

Australia, however, has many opportunities to capture this value and grow a battery recycling industry that is capable of processing the expected.[152] High volumes of battery waste are essential for the viability of recycling plants, and there are several enablers that could unlock this in Australia. Australia's policy environment is evolving and may result in greater recoveries of lithium battery waste for processing. For example, Victoria has banned disposing of lithium batteries in landfill, and other states are considering strengthening regulations.[153] Australia may need to consider importing battery waste from abroad, from particularly Asia and Pacific Islands. A 2021 CSIRO report discusses the regulatory landscape required to enable the creation of a regional recycling hub, including the key issue of transport costs.[154]

This section discusses the opportunities for domestic RD&D and for international collaboration in recycling technologies (summarised in Figure 8). For details on the framework used, refer to the main report.

[149] The $3 billion figure represents the lost/recoverable value from LiB wastes in 2036, based on 2021 commodity price; Zhao et al. (2021) Australian landscape for lithium-ion battery recycling and reuse in 2020 - current status, gap analysis and industry perspectives. CSIRO and FBI CRC; King S and Boxall NJ (2019) Lithium battery recycling in Australia: defining the status and identifying opportunities for the development of a new industry. Journal of Cleaner Production 215, 1279–1287. DOI: 10.1016/j.jclepro.2019.01.178.

[150] Terzon E (2023) EV batteries pose big risks – and new figures reveal how much hazardous waste they could create. <https://www.abc.net.au/news/2023-06-01/electric-vehicle-battery-waste-projections-uts-research/102417114>

[151] CSIRO (n.d.) Lithium-ion battery recycling. <https://www.csiro.au/en/research/technology-space/energy/energy-in-the-circular-economy/battery-recycling>

[152] CSIRO (n.d.) Lithium-ion battery recycling. <https://www.csiro.au/en/research/technology-space/energy/energy-in-the-circular-economy/battery-recycling>

[153] EPA Victoria (2023) Batteries. <https://www.epa.vic.gov.au/for-business/find-a-topic/manage-e-waste/batteries#:~:text=Batteries%20are%20an%20'electronic%20waste,a%20'specified%20electronic%20waste'>; Skatssoon J (2023) NSW considers banning lithium batteries from landfill. Government News. <https://www.governmentnews.com.au/nsw-considers-banning-lithium-batteries-from-landfill/#:~:text=Lithium%20batteries%20could%20be%20banned,the%20quest%20for%20renewable%20energy>

[154] King S, Boxall NJ, Bhatt AI (2018) Lithium battery recycling in Australia. CSIRO, Australia.

Figure 8: Actions for Australian RD&D and international collaboration in lithium-ion battery recycling.

Opportunity area	Establish new capability in emerging technologies	Accelerate emerging tech and grow Australian IP	Pilot and scale-up Australian IP	Support commercial deployment of mature technologies
Applicable Technologies	• Bio-hydrometallurgy (metal recovery) • Direct recycling	• Graphite regeneration • Electrolyte recovery • Electrochemical separation (metal recovery)	• Dismantling and separation • Pyro- and hydrometallurgy (metal recovery)	• Dismantling and separation • Pyro- and hydrometallurgy (metal recovery) • Discharging
RD&D actions	Build capability in emerging technology areas via fundamental and applied research projects.	Leverage Australia's strengths to progress technologies beyond the lab and grow Australian IP.	Deploy Australian IP in pilot-scale and commercial-scale demonstrations.	Support the deployment of mature technologies domestically at commercial scale, through commercial testing and validation, and cross-cutting RD&D.
International engagement actions	Engage with research institutions on capability building and knowledge sharing e.g. joint research programs	Partner with overseas industry, research or government on mutually beneficial sustained technology development efforts e.g. co-funded or joint projects.	Engage with upstream offtakers to de-risk and finance pilot projects. Alternatively, demonstrate Australian technologies overseas.	Engage on commercial arrangements e.g. international technology providers, license overseas patents, attract foreign direct investment, and secure offtake agreements.

IP, intellectual property. IP, intellectual property. For a full description and methodology of this framework, refer to the main report *From minerals to materials: Assessment of Australia's critical mineral mid-stream processing capabilities.*

The economics of recycling technologies must be examined in terms of end-to-end processes in the Australian context, as opposed to individual processing steps. For example, cutting costs in early processes could lead to higher costs in downstream processing, or an inability to recover high quality materials. Cross-cutting research, such as techno-economics is required to identify cost-effective, high recovery pathways, in light of Australia's current recycling infrastructure and systems, it's geography, projected battery uptake, policy, regulations and standards. In addition to recycling technology development, establishing a thriving battery recycling industry is dependent upon the effective gathering and sorting of spent LIBs to enable effective recycling.[155] Cross-cutting research is required to support the shift toward onshore recycling of LIBs, particularly in the areas of collection systems, harmonisation of standards, materials tracking, regulations for waste disposal and product stewardship.[156] Once collected, identified, sorted and stored appropriately, processing facilities and technologies focused on throughput can extract a range of material grades.

3.6.1 Discharging

Discharging is becoming increasingly important with higher volumes of large batteries entering waste streams (e.g. electric vehicle and stationary lithium ion batteries). This is essential to ensure the safety of workers and to protect investments made in high-tech recycling infrastructure. Further, updating discharging practices will be required to ensure that they are safe, and that high quality metals and materials can be recovered downstream and not contaminated or lost (as is the case with wet chemical discharging). Given Australia already undertakes disassembly and crushing prior to shipping black mass, there is a need to consider discharging technologies within Australia's recycling infrastructure.

Over the last 15 years, IP output in discharging technologies have been relatively low, suggesting that most technologies are mature and can leverage off the shelf equipment. The low levels of RD&D activity and

[155] Qadir et al. (2023) Formal and informal E-waste recycling methods for lithium-ion batteries: advantages and disadvantages. In Global E-Waste Management Strategies and Future Implications, 73-104, Elsevier.

[156] King S, Boxall J (2019) Lithium battery recycling in Australia: defining the status and identifying opportunities for the development of a new industry. Journal of Cleaner Production 215, 1279-1287.

industry uptake in discharging technologies, in Australia and globally, indicates there is an opportunity to build greater domestic capability via RD&D and international capability building efforts. Given the saturated IP landscape, RD&D opportunities are likely to be found in innovative solutions to facilitate commercial implementation and cross-cutting studies addressing regulatory aspects, rather than IP generation.

Opportunities for RD&D and collaboration include developing discharging systems that are compatible with mixed battery sizes and types, improving handling and thermal safety, reducing waste (for chemical methods), and enabling the recovery of discharged electricity for site or grid use.

3.6.2 Dismantling and separation

Dismantling, and separation is a necessary step in order to recover materials from lithium battery waste. Australia currently undertakes the dismantling and crushing aspects to produce black mass, and exports this for further processing. There is an opportunity for Australia to improve separation technologies to increase the quality of the back mass it exports to increase revenue.[157] Further, undertaking improved separation activities onshore this can unlock further domestic downstream processing activities.

Dismantling and separation is undertaken commercially at scale globally, however, research activity in this space has been strong with respect to emerging and improved techniques. This has been driven by a need to improve the effectiveness of binder removal in and enhance greater material recovery, as well as reduce cost, energy consumption and process waste. There are trade-offs with respect to physical, thermal and chemical separation methods or combinations of these. As such efforts are required to optimise processes that minimise cost, energy and reagent inputs, pollution, and the number of steps required, while enabling high-volume, high-quality material recovery downstream.

Despite limited domestic IP in this space, there is some opportunity for Australia to demonstrate innovative and improved separation and innovate in this domain to support downstream high-value metal recovery. There is also an opportunity collaborate with international partners that have commercial technology to support industry expansion, namely additional onshore separation activities to enable the integration of downstream recycling.

3.6.3 Graphite separation and regeneration

Graphite makes up to 22% of the mass of a LIB, but industrial processes and research have historically been focused on recovering the cathode material. Using mature processes, graphite is often unrecoverable and is discarded as waste or incinerated. With graphite shortages projected in the coming years, and given the finite nature of deposits and supply concentration of synthetic graphite, harnessing graphite from spent LIBs can ensure an additional supply source of low-cost graphite in the future.[158] Developing graphite regeneration methods is particularly important given graphite is the primary active anode material for LIBs across all cathode chemistries, including NCM, NCA and LFP batteries.[159] With regards to spent LFP batteries, regenerating high grade graphite may provide an additional revenue stream given high value metal recovery is lower than for other battery types.

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[157] Lim P (2022) 'Black mass' needs common global specs to commoditize recycled battery raw materials. <https://www.fastmarkets.com/insights/black-mass-needs-common-global-specs-to-commoditize-recycled-battery-raw-materials/>

[158] Onstad E (2023) Analysis: Auto firms race to secure non-Chinese graphite for EVs as shortages loom. Reuters. <https://www.reuters.com/world/china/china-require-export-permits-some-graphite-products-dec-1-2023-10-20>

[159] Syrah Resources Limited (n.d.) Battery Anode Market. <https://www.syrahresources.com.au/about/battery-anode-market>

The emergence of this technology area, growing momentum in global research, and Australia's research sector capability represents an opportunity to develop greater domestic IP. Graphite regeneration has not yet been widely adopted by battery recyclers, however, there has been a significant increase in the number of patents filed in this domain since 2017, indicating that importance of regenerating graphite is being recognised by industry and researchers alike. Australia has demonstrated capability in early stage research, ranking fifth in research publication output, however, has an absence of identifiable patent activity. In light of global momentum in graphite regeneration, Australia has an opportunity to develop technologies beyond the research lab and grow domestic IP for commercial application in domestic or international markets.

Developing processes that are able to yield battery grade graphite in cost effective, scalable and environmentally friendly ways is the key RD&D focus. Two key areas are enhancing pre-treatment steps to more effectively remove binders, and optimising hydrometallurgical purification processes, and reducing the energy intensity of thermal treatment steps.

3.6.4 Electrolyte recovery

Electrolyte recovery is often overlooked in LIB recycling processes, where it is typically allowed to evaporate and decompose during pre-treatment.[160] However, processes that ensure adequate, deliberate extraction are gaining interest in research and industry, with a sharp increase in patent activity since 2017. This is largely driven by the need to minimise the environmental pollution caused by the flammable and toxic materials present in the electrolyte, as well as economic benefits that may be captured. Additionally, separating and extracting electrolytes from battery shreds also yields a purer black mass, which is advantageous for further downstream processing to recover high value metals.[161] Further, integrating electrolyte recovery into current processes also enable recyclers to meet emerging global standards and demand for greater circularity.[162] For example, in 2023, new European Parliament regulation was introduced requiring that 50% of lithium is recovered from used batteries by 2027, and that new batteries must contain at least 6% of recycled lithium.[163]

Given Australia's world leading research output (top 5 globally) in electrolyte recovery and emerging IP activity, there is an opportunity to continue to grow domestic IP output and to commercially demonstrate exiting IP. RD&D areas include improving extraction efficiency and purity, minimising solvent use and fluorine gas emissions, and demonstrating Australian IP in domestic pilots or in collaboration with overseas battery or recycling stakeholders.

3.6.5 High value metal recovery

Worldwide, the economics of lithium battery recycling is dependent on the prices of high value metals as well as policy and regulatory incentives. The prices of CAM materials vary by type and by purity, for example in 2022, NMC material (high in nickel and cobalt) can be US$ 50 to 95 per kg, whereas LFP (low in

[160] Zachmann N, Petranikova M, Ebin B (2023) Electrolyte recovery from spent Lithium-Ion batteries using a low temperature thermal treatment process. Journal of Industrial and Engineering Chemistry, 118, 351-361.

[161] Stakeholder consultations.

[162] Zachmann N, Petranikova M and Ebin B (2023) Electrolyte recovery from spent Lithium-Ion batteries using a low temperature thermal treatment process. Journal of Industrial and Engineering Chemistry 118, 351–361. DOI: 10.1016/j.jiec.2022.11.020.

[163] European Parliament (2023) Making batteries more sustainable, more durable and better-performing. <https://www.europarl.europa.eu/news/en/press-room/20230609IPR96210/making-batteries-more-sustainable-more-durable-and-better-performing>

high value metals) can be worth US$ 35 to 50.[164] To date, operations have been primarily focused on the highest value revenue streams (e.g., nickel and cobalt). However, with growing market share of LFP batteries and general substitution towards lower cost battery chemistries, the economics of battery recycling could become more challenging. This is driving innovation in technologies that can increase revenue or enable new revenue streams for example, by recovering and valorising other materials, by overcoming material losses, or by lowering energy inputs.

As discussed above (Section 3.6.4), global policies are also pushing towards increased range, percentage and reuse rate of metals recovered from used batteries. These mechanisms are driving innovation in technologies and processes with comprehensive capabilities, and novel recycling strategies.

Pyrometallurgical and hydrometallurgical extraction

Pyrometallurgical and hydrometallurgical pathways are employed at large scale in industry globally, however, RD&D activity in these areas continues to experience high levels of growth, driven by a need to more effectively extract high quality materials at lower costs.

Australia possesses RD&D capability in high value metal recovery; however, activity is weighted more strongly in the early research stages rather than IP development and piloting. There is a potential for Australia to increase its translation of research into generation of IP in this area, or to engage internationally to adopt commercially mature solutions.

RD&D and collaboration opportunities exist across both pyrometallurgical and hydrometallurgical pathways. Pyrometallurgy (particularly smelting) is the most commonly employed method in industry globally, and therefore benefits from lower risk. However, it is less well suited to LFP chemistries, requires high temperatures, and leads to relatively large material losses including lithium, graphite, and polymers. Market shifts towards LFP batteries, mixed or complex battery feedstocks can render current smelting techniques uneconomical and vulnerable.[165] As such, there are several RD&D opportunities to improve pyrometallurgical pathways such as reducing energy intensity, improving recovery rates (such as recovering lithium from electrolytes or slags), and developing alternative pyrometallurgical methods such as roasting.

Hydrometallurgy is experiencing increased commercial deployment and interest across international and Australian industry proponents, due to its high recovery rates, high purity product, and its potential for processing low grade feedstocks. RD&D opportunities in this area relate to minimising the cost of reagents, chemical recycling, and optimising the process at scale.

Technology choice will depend on several complex factors including optimising the end-to-end process and the broader recycling ecosystem (including the development and harmonisation of battery standards, collection, transportation, and sorting). This requires further analysis and is not within the scope of this report. However, it represents an opportunity for enabling RD&D (see Synthesis Report).

Given the low volumes of domestic EV battery waste in Australia, there is likely a limited number of commercial pilots that can be done with a view to scale up. As such, Australia's RD&D investment will need to be targeted and international collaboration will play a strong role in knowledge sharing and mitigating duplication of effort. Demonstrations or small-scale operations may be built near Australian CAM and battery manufacturing plants. Taking in homogeneous feedstock from plant waste or battery stewardship programs, reduces the need for complex processing of mixed battery waste. Australia may need to

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[164] Kaya M (2022) State-of-the-art lithium-ion battery recycling technologies. Circular Economy. https://doi.org/10.1016/j.cec.2022.100015>

[165] Makuza et al. (2021) Pyrometallurgical options for recycling spent lithium-ion batteries: A comprehensive review. Journal of Power Sources.

collaborate internationally to demonstrate its IP in overseas markets with comparatively high levels of EV waste, and to license its technology overseas.

Emerging processes

Electrochemical separation processes have the potential to overcome the complexity and reagent intensity of conventional hydrometallurgical separation pathways, and they are being piloted globally. There is an opportunity to leverage Australia’s cross-cutting capabilities from other industries to develop relevant processes for recycling and potentially scale up beyond the lab. RD&D would be required to improve the selectivity and efficiency of the process. Partnering with emerging developers who are piloting this technology or engaging in joint research projects might be beneficial for knowledge sharing and capability building, ultimately increasing the technology’s global maturity.

The direct recovery of CAM represents a step change opportunity in terms of sustainability and cost, relative to incumbent pyrometallurgical and hydrometallurgical pathways. Direct recovery reduces the number of metal separation and purification steps required compared to pyrometallurgical and hydrometallurgical processes targeting separated metals. Given global capabilities in direct recycling are nascent, there is an opportunity for Australia to engage internationally to build its capability in this space.

Finally, bio-hydrometallurgical extraction is an emerging area globally, and is able to effectively recover high value metals from spent LIBs with advantages over other methods in terms of capital costs, energy consumption and environmental impact. Although bio-hydrometallurgical extraction has not been commercially applied on batteries, the technology is mature for the extraction of other metals from ores, from which knowledge, skills and lessons can be transferred.

As discussed, Australia is developing strong capability in its research sector for batteries and has cross-cutting industry and research capabilities in bio-extraction from e-waste and ores which can be leveraged for battery waste applications. This represents an opportunity for Australia to continue to build applied capability in the recycling domain, and to engage in capability building with international partners.

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