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TECHNICAL BRIEF

Novel Advances in Lithium Ion Battery Recycling and Pretreatment

INTRODUCTION

The use of lithium ion batteries (LIB) in electric vehicles, stationary energy storage, and consumer products continues to increase exponentially, and industry is preparing for a rising demand for end-of-life (EOL) management services. At the point when LIBs are no longer viable in the original product, the batteries can be reused, repurposed for use in another product type (e.g., electric vehicle batteries to stationary storage), recycled, or disposed of in a landfill.[1,2] LIB modules are usually classified as universal waste (a category of hazardous waste with simplified handling requirements). Landfilling – even in a Title C hazardous waste landfill – is not recommended because of the benefits associated with the other options. Recycling LIBs creates value out of what would otherwise be waste and avoids virgin material mining that would be required to manufacture new batteries. This avoids associated environmental, social, and global transport impacts from obtaining lithium, cobalt, nickel, and other metals. Collecting used modules is necessary to 1) establish domestic accessibility to the valuable and critical materials and 2) support the fledgling LIB recycling industry in many countries worldwide. As a result, recycling has emerged as an advantageous and recommended option for LIBs.[3] This white paper reviews the state of the LIB recycling industry, commercially available recycling methods, and promising recycling innovations and advancements that could be commercialized by 2030.

COMMERCIALLY AVAILABLE LIB RECYCLING METHODS

EOL batteries may be discharged as a first step in the recycling process, but certain methods do not require it. The modules are disassembled to separate cells and remove casings and connectors. Cells may be sorted to allow for different treatment by cathode chemistry to maximize metal recovery. The cells and modules are then shredded and crushed, usually in an inert atmosphere or in an aqueous bath to minimize risks of fire. Plastic, metal foil, and other materials are separated based on size, density, or other properties. The resulting black mass contains the valuable LIB materials that must be extracted through pyrometallurgy, or hydrometallurgy.

Pyrometallurgy relies on high-temperature smelting to burn off non-metals and isolate the metallic elements as alloys. This method offers several advantages: pyrometallurgy reduces or eliminates the need for sorting, size reduction, and pretreatment, and has been commercially mature for longest. However, pyrometallurgy requires a large amount of energy achieve the requisite high temperatures and produces substantial greenhouse gas and air pollutant emissions. Additionally, some of the desired constituents may be lost in the process and further treatment of the alloys is necessary to extract the individual metallic elements.

1 *Guidelines for Assessing End-of-Life Management Options for Renewable and Battery Energy Storage Technologies*. EPRI, Palo Alto, CA: 2021. 3002020594.

2 *Second Life for Large-Format Lithium Ion Batteries*. EPRI, Palo Alto, CA: 2023. 3002028618.

3 *End-of-Life Management for Lithium Ion Battery Technologies: Issues, Uncertainties, and Opportunities*. EPRI, Palo Alto, CA: 2021. 3002020006.

Hydrometallurgy applies chemical separation methods to selectively dissolve and isolate the metallic elements. This method isolates most constituents at high purity, but is also costly and complex, as it requires sorting, size reduction, multiple chemical separation processes, and wastewater management. Hydrometallurgy has emerged in the last couple years as a commonly used commercially available method, and there are now many commercial hydrometallurgical recycling facilities in operation.[4] NAATBatt's Lithium-ion Supply Chain Database[5] shows that 12 out of the 21 recycling facilities that are under construction or planned will be using hydrometallurgy, though not all facilities in the database specify process details.

VENDOR LANDSCAPE

In-depth profiles of several lithium ion battery recyclers are provided to explore the range of process approaches commonly being taken in the industry and the extent of publicly shared details.

Redwood Materials

Redwood Materials is based in Carson City, Nevada, and was founded in 2017. They have one operational battery recycling facility in McCarran, Nevada and are building a second facility in Charleston, South Carolina.

Redwood's recycling method utilizes three refinement processes, including reductive calcination, mechanical processing, and hydrometallurgical processing.[6] Energized battery feedstock is first pre-processed through reductive calcination, which leverages the exothermic reactions of the battery materials to drive the thermal process instead of an external energy source. The reducing conditions also minimize combustion of carbon materials, which in turn reduces CO_2 emissions. The calcined materials are then refined through mechanical processing to separate various alloys.

The final step is a proprietary hydrometallurgical process, where the inputs are turned into a slurry and pumped into acid leaching tanks. Metals are dissolved into the solution with sulfuric acid and other reagents, and the graphite is removed through a filter press. The metal sulfates-rich filtrate undergoes downstream processing for separation and purification. No hydrogen peroxide (typically used as a reducing agent for transition metals) is used in Redwood's hydrometallurgical process; it is unknown if an alternate agent is used or if the thermal reduction accomplishes that goal. Redwood states that it plans to use reclaimed water and will recycling approximately 90% of all water used on site.[7] Redwood claims a 95% recovery rate of lithium.[8]

Redwood intends to build a cathode active material (CAM) facility to process the metal salts formed in the hydrometallurgical step. Argonne National Laboratory has validated the performance of NMC-811 cathodes using pCAM (CAM precursor) from Redwood's recycling process, finding that the cathode materials made from the metal sulfates produced from recycled batteries had similar cycle life, discharge capacity, and Coulombic efficiency as those made from virgin mined metals.[9] Redwood produces pCAM through co-precipitation of the metal salts, and then combines it with lithium salts and other additives. After being oxidized, this product is milled, classified by size, and dried to produce CAM.

Redwood is also building a copper-foil facility for anode current collectors. The copper feedstock generated by the hydrometallurgical process is dissolved in acidic solution and then electrodeposited upon a metal drum. This creates a thin copper foil that can be peeled off and stored in rolls.

4 Davis, K. and Demopoulos, G.P. Hydrometallurgical recycling technologies for NMC Li-ion battery cathodes: current industrial practice and new R&D trends. *Royal Society of Chemistry*. https://pubs.rsc.org/en/content/articlehtml/2023/su/d3su00142c.

5 National Renewable Energy Laboratory. NAATBatt Lithium-Ion Battery Supply Chain Database. 2024. https://www.nrel.gov/transportation/li-ion-battery-supply-chain-database.html.

6 Machala, Michael and Chen, Xi and Bunke, Samantha P. and Forbes, Gregory and Yegizbay, Akarys and de Chalendar, Jacques and Azevedo, Ines and Benson, Sally M. and Tarpeh, William, Life Cycle Comparison of Industrial-Scale Lithium-Ion Battery Recycling and Mining Supply Chains. Available at SSRN: https://ssrn.com/abstract=4309094 or http://dx.doi.org/10.2139/ssrn.4309094.

7 U.S. Department of Energy. *Environmental Assessment: Construction of Production Facilities for High-Quality Cathode Active Materials and High-Performance, Ultra-Thin Battery Copper Foil*. https://www.energy.gov/sites/default/files/2023-11/231128_Final_Redwood_EA_FONSI.pdf.

8 Redwood Materials. "Building the most sustainable (and scalable) battery materials process." https://www.redwoodmaterials.com/news/sustainable-battery-materials-process/.

9 Redwood Materials. "U.S. Department of Energy's Argonne National Laboratory verifies performance of Redwood cathode from recycled content." October 13, 2022. https://www.redwoodmaterials.com/news/argonne-national-laboratory-verifies-redwood-cathode-performance/.

Southern Company.[10] This was Redwood's first standalone BESS decommissioning project, as their prior experience focused on receiving, processing, and recycling battery materials. During the decommissioning, Redwood was responsible for de-energizing, disassembly, packaging, shipping, and recycling. Redwood completed the recycling of these batteries in 2023.

Li-Cycle

Established in 2016, Li-Cycle is headquartered in Toronto, Canada, and has multiple battery recycling facilities located in the Greater Rochester Area, New York, commissioned in 2020; Gilbert, Arizona, commissioned in 2022; Tuscaloosa, Alabama, commissioned in 2022; and Magdeburg, Germany, commissioned in 2023.[11] As of August, 2024, Li-Cycle closed its first battery recycling facility located in Kingston, Ontario, which had been established in 2019.[12] Further expansion plans in New York, France, Italy, and Norway have been paused due to higher than expected construction costs.

Li-Cycle refers to its mechanical processing facilities as "Spokes" and hydrometallurgical facilities as "Hubs". All operational facilities are Spokes, and construction for the first planned Hub is currently paused. Li-Cycle's recycling process begins with submerged shredding of batteries and scrap. Batteries of any format, chemistry, or state of charge can be processed in this step to be turned into inert materials. Further mechanical processing separates the black mass containing the battery materials from the mixed copper and aluminum product. Li-Cycle plans to then transport the black mass to a centralized Hub, where the black mass will be processed through a hydrometallurgical step to produce battery materials. The Spoke recycling efficiency was independently validated at 73.8% in 2021 based on the now-closed Ontario Spoke, and Li-Cycle currently claims a 95% recovery rate based on further improvements.[13]

Li-Cycle has made a 2023 Sustainability Report[13] publicly available, and select results from Li-Cycle's independently conducted LCA are included. Waste stream monitoring revealed that of the waste generated by the North American Spokes and warehouses in 2023, 84% of materials by mass (8,202 tonnes) were diverted from landfill and 16% was landfill waste (1,610 tonnes). Most of the materials diverted from landfill consisted of black mass product (4,324 tonnes), shredded metal by-product (2,748 tonnes), and scrap metals from pre-processing (1,017 tonnes). Li-Cycle expects no direct emissions from the submerged shredding process at Spokes, but Scope 1 emissions from its fleet vehicles remain. Li-Cycle has electrified 81% of its fleet, mostly electric forklifts. For water management, Li-Cycle intends to reuse process water in a closed-loop system to minimize wastewater discharge and water demand. In 2023, Li-Cycle used 12625 m^3 of water for all Spokes (excluding the Rochester Hub and Germany Spoke).

American Battery Technology Company (ABTC)

ABTC was founded in 2011 and is headquartered in Reno, Nevada. ABTC currently has one battery recycling facility, also located in Reno. ABTC also has a location in Nevada developing primary extraction processes for responsible, sustainable mining.

ABTC's recycling process begins with an automated "de-manufacturing" process, where batteries are disassembled into cell and sub-cell components. The batteries may still have residual energy, which must be managed. Copper, aluminum, steel, a lithium intermediate, and a black mass intermediate are separated. Through hydrometallurgical processes such as acid leaching with a reducing agent, the lithium intermediate is refined to lithium hydroxide, and the black mass intermediate is refined into nickel, cobalt, manganese, and lithium hydroxide products. ABTC does not rely on any high temperature operations, and no combustion occurs during any of its recycling processes. ABTC claims a recovery rate of 90%.

10 *Cedartown Battery Energy Storage System Decommissioning Case Study: Lessons Learned from Decommissioning an Early-Stage Utility-Scale Lithium Ion Project*. EPRI, Palo Alto, CA: 2023. 3002027944.

11 Li-Cycle. "Spoke & Hub Network." https://li-cycle.com/operations/.

12 Stafford, T. Employees suddenly laid off at Li-Cycle Kingston facility. *Kingstonist*, November 2023. https://www.kingstonist.com/news/employees-suddenly-laid-off-at-li-cycle-kingston-facility/.

13 Li-Cycle. *2023 Sustainability Report Environment, Social, and Governance Performance*. 2023. https://li-cycle.com/wp-content/uploads/2024/05/2023-Li-Cycle-Sustainability-Report.pdf.

RECYCLING INNOVATIONS AND ADVANCEMENTS

The following section is an overview of promising innovations and advancements in the lithium ion battery recycling space, including improvements to existing processes, novel methods, and reducing risk during transport of batteries to a recycling facility.

Improvements to Existing Processes

Hydrometallurgy is a part of many companies' recycling processes. Metal or metal oxide recovery rates are commonly in the 90-98% range, making additional recovery efficiency improvements difficult. Thus, research focus has been on optimizing high value by-product production or reducing waste streams.

For example, one drawback of hydrometallurgical lithium ion battery recycling is that it produces sodium sulfate as a byproduct, a waste stream with few options for disposal.[14] Sodium sulfate can be pumped into the ocean or sold to dry laundry detergent companies that use sodium sulfate, but sodium sulfate production is expected to increase as the industry grows. The detergent industry, especially outside of China, is not projected to be able to make use of all of the sodium sulfate, and disposal into rivers and oceans has prompted backlash from communities and environmental agencies.[15] Several companies are developing solutions to the sodium sulfate waste stream. Cinis Fertilizer has partnered with Northvolt, a battery manufacturer with a sodium sulfate byproduct, to convert the sodium sulfate into potassium sulfate fertilizer.[16] Aepnus Technology is converting sulfate salts into high concentration sulfuric acid and metal hydroxides that can be used by the battery industry.[17]

Other hydrometallurgical LIB recycling process improvements being researched include reducing acid and base consumption; using alternate reagents such as KOH instead of caustics; using regenerative lixiviants such as HCl or HNO_3 instead of H_2SO_4 to recycle the solvents; efficient fluoride removal; and advanced process controls to help remove impurities through continuous sensing.[18]

Ascend Elements[19] is commercializing a proprietary Hydro-to-Cathode® direct precursor synthesis process to improve upon traditional hydrometallurgy. The batteries undergo de-energization, shredding and acid leaching. However, unlike typical hydrometallurgy processes where the critical materials are individually extracted, Ascend Elements extracts impurities like aluminum, copper, plastics, and graphite, and leaves the nickel, cobalt, and manganese in solution. During the elemental adjustment step that follows, the type of pCAM or CAM can be specified in solution, including the metal ratios, particle size, crystallinity, porosity, and morphology of the cathode. The cathode precursor is then synthesized and precipitated out. Ascend Elements' first commercial pCAM manufacturing plant is under construction in Hopkinsville, KY. It is expected to come online in 2025 and will have the capacity to produce 27 kT of pCAM per year in 2026.[20] Ascend Elements has also released select results from an independently conducted LCA, which found that carbon emissions from the Hydro-to-Cathode® process are 21.9 kg per kg of cathode, 49% lower than producing a cathode with primary materials from mining.[21]

Other companies are pursuing advanced feed management systems, including the use of robotics, artificial intelligence (AI), and machine learning (ML) to reduce risks to human workers and reduce the cost of recycling. Universe Energy[22] plans to utilize robotics to disassemble modules, with a software that proposes a disassembly sequence for each module type. The Swiss Battery Technology Center

14 Blois, M. What to do with the battery industry's sodium sulfate waste?. *Chemical & Engineering News*, Volume 102, Issue 4, 2024. https://cen.acs.org/energy/battery-industrys-sodium-sulfate-waste/102/i21.

15 Blois, M. BASF battery project delayed because of environmental concerns: Other firms building battery materials plants in the US and Europe could face similar permitting hurdles. *Chemical & Engineering News,* Volume 102, Issue 7, 2024. https://cen.acs.org/environment/pollution/BASF-battery-project-delayed-environmental/102/i7.

16 Cinis Fertilizer. "Circular, sustainable and cost-efficient production process." https://www.cinis-fertilizer.com/what-we-do/production-process/.

17 Aepnus Technology. "A New Breed Of Electrolysis Technology." https://aepnus.com/technology.

18 Anson, James, "Accelerating Innovation," NAATBatt Lithium Battery Recycling Workshop VII: Closing the Circle. Montreal, Quebec (August, 2024).

19 Ascend Elements. "Sustainable lithium-ion battery materials and recycling — elevated." https://ascendelements.com/.

20 Ascend Elements. "New Visions of Our Apex 1 Campus." https://ascendelements.com/new-visions-of-our-apex-1-campus/.

21 Duhayon, C., Ph.D.; Garaycochea, A, M.S.; and Giddings, E. B.S. *The Road to Net Zero: Summary of life cycle assessment for NMC 622 CAM products made from recycled lithium-ion batteries via the Hydro-to-Cathode® process.* Ascend Elements, January 2024. https://ascendelements.com/wp-content/uploads/2024/01/AE-LCA-White-Paper-2024-FINAL.pdf.

22 Universe Energy. "The battery dismantling and sorting company." https://universeenergy.ai/.

has partnered with two companies, Circu Li-ion and AICA SA, to use AI and ML to teach robots how to dismantle a variety of battery types as well as damaged batteries.[23] Oak Ridge National Laboratory[24] demonstrated an automated disassembly system at lab-scale in 2021, where robots skip the de-energizing of the battery and begin physical disassembly that can be configured to access only the individual modules for refurbishing or reuse, or configured to break down the battery to the cell level for recycling. A literature review[25] of robotized disassembly of EV batteries found that separation, testing, sorting, cutting, grasping, detecting, and unscrewing have been investigated in publications as a potential task that could be automated. The proposed solutions typically relied on machine vision to locate and classify, an arm robot to physically interact with the modules, and AI to control the disassembly process. However, challenges remain with the design of batteries: the lack of standardization complicates automation, and the use of permanent joints, like welding and non-reversible adhesives, requires a destructive separation method. To bypass these challenges, human-robot collaboration is used to complete tasks that require cognitive abilities and finger dexterity that are beyond the current capabilities of robotics.

Novel Recycling Methods: Direct Recycling

Several companies are investigating direct recycling[26] as an alternative battery recycling method. With currently utilized methods like pyrometallurgy and hydrometallurgy, the end-of-life battery is broken down to the raw materials used for manufacturing. Direct recycling would isolate and preserve the structure of the cathode, the costliest component of a battery cell, reducing manufacturing cost and waste and retaining the embedded energy used to create the initial cathode.

OnTo Technology[27] is commercializing a hydrothermal relithiation method for direct recycling called cathode-healing™. OnTo Technology demonstrated this method as a part of the full direct recycling process, starting with a feedstock of recalled LCO batteries and ending with the manufacturing of lithium ion cells with the recycled materials.[28] The process begins with discharge of the batteries through a sodium bicarbonate bath and the extraction of the electrolyte from the batteries with a CO_2 extraction system. The cells are shredded and filtered to separate plastics, metal casings, and current collectors from the electrode material, which contains cathode, carbon black, binder, and graphite. The electrode material undergoes hydrothermal treatment with a lithium aqueous solution, which relithiates the spent cathode material and removes the binder, impurities, and trace electrolyte. The cathode is isolated through froth floatation and heated, while the recovered graphite is purified. OnTo built and characterized half-cells and full cells using recycled graphite and cathode material, and the cells with recycled cathode material demonstrated a return to baseline capacity and power.

Princeton NuEnergy[29] is developing another direct recycling method using Low-Temperature Plasma Assisted Separation, or LPAS™. PNE's process begins with shredding of battery waste and manufacturing scrap, and then separates the cathode and anode materials. The cathode materials undergo plasma purification through LPAS™, producing the cathode active material precursors (pCAM). Princeton NuEnergy utilizes a Micro-Molten Shell-Assisted Lithiation (MSAL™) process to relithiate and restore the structure of the cathode materials. Princeton NuEnergy produces CAM, purified anode materials, and advanced black mass (ABM™), and additionally separates copper, aluminum, plastics, and electrolyte for use and recycling by third parties.

23 GGB[a]. "Swiss Battery Technology Center innovates with robotic battery recycling." Invest in Western Switzerland. https://ggba.swiss/en/swiss-battery-technology-center-innovates-with-robotic-battery-recycling/.

24 Oak Ridge National Laboratory. "Automated disassembly line aims to make battery recycling safer, faster." *ORNL News*. https://www.ornl.gov/news/automated-disassembly-line-aims-make-battery-recycling-safer-faster.

25 Kaarlela, T.; Villagrossi, E.; Rastegarpanah, A.; San-Miguel-Tello, A.; and Pitkäaho, T. Robotised disassembly of electric vehicle batteries: A systematic literature review. *ScienceDirect*, Volume 74, June 2024, Pages 901-921. https://doi.org/10.1016/j.jmsy.2024.05.013.

26 Wang,Y.; Zhai, Q.; and Yuan, C. Analysis of Direct Recycling Methods for Retired Lithium-ion Batteries from Electric Vehicles. *ScienceDirect*, Volume 116, 2023, Pages 702-707. https://doi.org/10.1016/j.procir.2023.02.118.

27 Onto Technology. "Leader in closed-loop lithium-ion battery technology." https://www.onto-technology.com/.

28 Sloop, S.; Crandon, L.; Allen, M.; Koetje, K.; Reed, L.; Gaines, L.; Sirisaksoontorn, W.; and Lerner, M. A direct recycling case study from a lithium-ion battery recall. *ScienceDirect*, Volume 25, September 2020, e00152. https://doi.org/10.1016/j.susmat.2020.e00152.

29 Princeton NuEnergy. "Lithium-ion Battery Direct Recycling." https://pnecycle.com/.

Safer Transport of Batteries for Recycling

LIB EOL management encompasses more than the recycling processes. One of the most expensive parts of decommissioning a stationary energy storage system is transportation of batteries to the recycling facility due to the ignition risk. Innovations reducing this risk may have positive cost and safety implications. Battery modules that may be damaged, defective or recalled (DDR) also pose unique risks that must be managed during transport.[30]

KULR[31] is a company that is developing packaging technology for the safer transport of lithium ion batteries. KULR's passive propagation resistant packaging includes a Thermal Runaway Shield (TRS) that can prevent cell-to-cell propagation. The TRS was tested for use on the International Space Station and was found to prevent thermal runaway in laptop computer battery packs that were tightly packed in fire-proof bags.[32] The TRS is incorporated into KULR's SafeCASE and SafeSLEEVE products, which are intended for the safe storage and transportation of batteries by providing physical and thermal protection.[33] These products are approved by the US Department of Transportation (DOT) for ground transportation and can hold 2.5 kWh and 0.3 kWh respectively.[34] KULR recommends the 2.5 kWh-sized product for lithium ion battery transportation and storage, including for DDR batteries. The 0.3 kWh-sized product fits small portable devices, including laptops, cellphones, power banks, and portable gaming devices. KULR intends to scale up its products to increase the capacity of the battery that can be accommodated.

The Pipeline and Hazardous Materials Safety Administration (PHMSA) is a US DOT agency, responsible for ensuring the safe transportation of energy and hazardous materials. PHMSA is developing packaging and safety solutions for DDR batteries, which may also be applicable for non-DDR batteries. While still under development, PHMSA's solutions include: smart drums that can contain battery fires and neutralize generated gases; an ISO/Conex container with plug-in fire suppression and ventilation; and a logistics trailer with active fire suppression and explosion, ventilation, and circulation systems.[30]

The previously mentioned company OnTo Technologies is also developing a method to neutralize lithium ion battery modules onsite at a storage facility or fleet storage location. The batteries are neutralized through CO_2 extraction, where active lithium is irreversibly eliminated using supercritical CO_2. This process results in a product that is a neutral solution with no flammability, toxicity, or arc hazard, while preserving the cathode materials. OnTo Technologies has demonstrated this deactivation method at their pilot plant and claims to eliminate all class-9 hazards in lithium ion batteries.[35]

CONCLUSION

Lithium ion battery recycling is an emerging industry with many companies seeking to scale up and commercialize their processes and offering new innovations to improve the safety, recovery rate, and cost of recycling. Companies closest to commercialization are primarily relying on hydrometallurgy, while other companies are working on direct recycling in an effort to leapfrog them due to reduced energy needs and cost for the same amount of battery processing. Continued improvements are necessary in not only the recycling process itself, but also in reducing the risks of transporting end-of-life battery modules for recycling, especially if those modules are damaged. Continued advancements in lithium ion battery transportation, pretreatment and recycling will be crucial for managing the increasing volume of end-of-life batteries in the most safe and economically and environmentally advantageous manner.

30 *Decommissioning Strategies for Damaged, Defective and Recalled Lithium Ion Batteries*. EPRI, Palo Alto, CA: 2024. 3002031225.

31 Kurl Technology. "Building an Energy Management Platform." https://www.kulrtechnology.com/.

32 Kurl Technology. "KULR Technology Group Announces Partnership with Leidos to Supply NASA with Lithium-Ion Battery Storage Solution for International Space Station." https://www.globenewswire.com/news-release/2019/09/03/1910073/0/en/KULR-Technology-Group-Announces-Partnership-with-Leidos-to-Supply-NASA-with-Lithium-Ion-Battery-Storage-Solution-for-International-Space-Station.html.

33 Kurl Technology. "Thermal Solutions for Lithium-Ion Battery Safety." SafeX. https://www.kulrtechnology.com/safe-case/.

34 Kurl Technology. "Battery Storage and Transportation Solutions." SafeX PRODUCTS. https://www.kulrtechnology.com/wp-content/uploads/2023/01/SafeCase-Sell-Sheet-1.pdf.

35 Onto Technology. "Products & Services." https://www.onto-technology.com/products-services.

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