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**Development of Efficient and Scalable
Direct Recycling Technology for
Lithium-Ion Batteries**

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PREFACE

The California Energy Commission's (CEC) Energy Research and Development Division supports energy research and development programs to spur innovation in energy efficiency, renewable energy and advanced clean generation, energy-related environmental protection, energy transmission, and distribution and transportation.

In 2012, the Electric Program Investment Charge (EPIC) was established by the California Public Utilities Commission to fund public investments in research to create and advance new energy solutions, foster regional innovation, and bring ideas from the lab to the marketplace. The EPIC Program is funded by California utility customers under the auspices of the California Public Utilities Commission. The CEC and the state's three largest investor-owned utilities—Pacific Gas and Electric Company, San Diego Gas and Electric Company, and Southern California Edison Company—were selected to administer the EPIC funds and advance novel technologies, tools, and strategies that provide benefits to their electric ratepayers.

The CEC is committed to ensuring public participation in its research and development programs that promote greater reliability, lower costs, and increase safety for the California electric ratepayer and include:

- Providing societal benefits.
- Reducing greenhouse gas emission in the electricity sector at the lowest possible cost.
- Supporting California's loading order to meet energy needs first with energy efficiency and demand response, next with renewable energy (distributed generation and utility scale), and finally with clean, conventional electricity supply.
- Supporting low-emission vehicles and transportation.
- Providing economic development.
- Using ratepayer funds efficiently.

EPC-21-008 is the final report for the *Development of Efficient and Scalable Direct Recycling Technology for Lithium-Ion Batteries* project conducted by the University of California, San Diego. The information from this project contributes to the CEC Energy Research and Development Division's EPIC Program.

For more information about the Energy Research and Development Division, please visit the [CEC's research website](http://www.energy.ca.gov/research/) (www.energy.ca.gov/research/) or contact the Energy Research and Development Division at ERDD@energy.ca.gov.

ABSTRACT

The rapid accumulation of end-of-life lithium-ion batteries poses a critical challenge, necessitating efficient recycling to preserve the environment and ensure a sustainable supply of critical materials. This project focused on scaling up direct recycling technology originally developed at the University of California, San Diego, to a multi-kilogram pilot scale under representative operational conditions. Key objectives were achieved, including the refinement of de-energization processes for electric vehicle batteries with varying chemistries, enhancement of cathode and anode material separation processes to achieve over 95 percent purity, over 90 percent yield of active material retrieval from spent batteries, and successful regeneration of 5 kilograms of active material through an energy-efficient recycling process. New lithium-ion battery cells were fabricated from recycled active material and demonstrated a performance equivalent to that of cells made from virgin materials. Above 99-percent purity of cathodes and 99-percent capacity retention as compared to virgin materials was achieved. Life-cycle analysis further evaluated the energy, economic, and environmental impacts of the entire process compared with conventional recycling. Future projects will aim to refine protocols to enhance energy efficiency, reduce operational costs and processing time, and scale cathode regeneration to 100 kilograms per day, with a parallel emphasis on anode regeneration. These results lay the foundation for future commercialization efforts to establish direct recycling capabilities in California.

Keywords: Lithium-ion batteries, direct recycling, hydrothermal, cathode, scaling

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TABLE OF CONTENTS

Acknowledgements	i
Preface.....	ii
Abstract	iii
Executive Summary.....	1
Project Purpose and Approach	1
Key Results.....	2
Knowledge Transfer and Next Steps.....	3
CHAPTER 1: Introduction	4
CHAPTER 2: Project Approach	7
Battery Acquisition, Diagnosis, Sorting, and De-energization.....	8
Cathode and Anode Materials Separation.....	10
Improve Efficiencies of Direct Recycling Processes	13
Cathode Recovery	13
Anode Recovery	17
Direct Recycling Process Scale-up	19
Performance Evaluation and Demonstration of Direct Recycling of LIBs.....	21
CHAPTER 3: Results and Discussion	23
CHAPTER 4: Technology and Knowledge Transfer.....	33
CHAPTER 5: Conclusion.....	36
Benefits to Ratepayers	36
Potential of Demonstrated Technology	37
Need for Future Support.....	37
Future Research Recommendations	38
Glossary and List of Acronyms	39
References	42
Project Deliverables.....	44
APPENDIX A: Relevant Protocols, Calculations, Materials Structure, Composition, and Electrochemical Property Data	A-1

LIST OF FIGURES

Figure 1: Simplified Schematic Comparison for Recycling Processes	5
Figure 2: Illustration of Technology and Its Advantages	6

Figure 3: Battery Modules Collected From Various Sources	9
Figure 4: Kilogram Level Black Mass Recovery for Scaling up Regeneration	11
Figure 5: Separation Workflow From Shredding to Froth Floatation	12
Figure 6: Froth Floatation Separation and Purity Analysis of the Product.....	12
Figure 7: Schematic of the Process Indicating Scalable Regeneration of CAM by Multistep Integrated Direct Recycling.....	14
Figure 8: Illustration of PVDF Decomposition Mechanism During PRIME Process	15
Figure 9: FTIR Spectroscopy Analysis of Decomposed Binder (PVDF)	16
Figure 10: (a) Solution Reuse Strategy Schematic and (b) Electrochemical Performance of the Recycled Product - Generation 2 (rCAM-G2).....	16
Figure 11: Purified Graphite Product Obtained After Recycling; Evident From (a-b) SEM and Respective (c-d) EDS* Data	17
Figure 12: Electrochemical Performance of Recovered Graphite Product (rAAM) Tracked at Each Regeneration Step	18
Figure 13: Schematic for the ABM Regeneration to Upcycled Graphite.....	19
Figure 14: Typical Impurities to Deal With After Sieving (<100 microns) to Obtain Feedstock for Direct Recycling	20
Figure 15: Preprocessing Schematic to Obtain CBM From the Battery Modules Obtained From Spent EV Packs Received From Vendor B	21
Figure 16: Morphology and Electrochemical Performance of the Recycled LCO Sample From LIBs.....	24
Figure 17: Crystal Structure Evolution of Recycled NCM111 CAM (rCAM) from CBM Compared to Pristine NCM111 Material (pCAM).....	25
Figure 18: Impurity Analysis of (a) Carbon by Combustion Analysis and (b) Aluminum by EDS, for Quality Comparison Between Recycled NCM111 CAM (rCAM) and Pristine NCM111 Material (pCAM).....	25
Figure 19: XPS* Data of (a) Fluorine, (b) Oxygen and (c) Carbon to Indicate Removal of the Binder and Cathode Electrolyte Interphase From the Recycled Cathode (rCAM).....	26
Figure 20: Morphology and Electrochemical Performance of the Recycled NCM111 Sample From Spent LIBs	26
Figure 21: Electrochemical Performance of the Recycled NCM111 CAM (rCAM) and Pristine NCM111 Material (pCAM): a) Rate Performance of the Regenerated Cathode Compared With Pristine, and b) Cycling Stability in Full Cell	27
Figure 22: Electrochemical Performance of the Recovered NCM622 Cathode (rCAM622) and Control CBM622: (a) First-cycle Voltage Profile and (b) Cycling Stability.....	28

Figure 23: Comparison of Various Recycling Method Calculated Using Everbatt Model	29
Figure 24: Scaled-up Feedstock Recovery From Spent EV Battery Packs Using the Pretreatment Protocol Developed in This Project.....	30
Figure 25: Demonstration of Direct Recycling Scale-up via the PRIME Process Developed in This Project.....	30
Figure 26: Pouch Cell Testing of rCAM NCM111 for Electrochemical, Power, and Impedance Performance Verification	31
Figure 27: HPPC Testing Protocol Used to Test Impedance and Power Performance of Pouch Cells.....	32
Figure 28: Scaled Preprocessing Equipment.....	34
Figure 29: Scaled-up PRIME Reactor Size	34
Figure 30: Scaled-up Post-PRIME Processing	35
Figure A-1: Battery 1 Pulse Discharge Profile	A-2
Figure A-2: Battery 2 Pulse Discharge Profile	A-3
Figure A-3: Instrument and Setups used for Diagnosis and De-energizing	A-4

LIST OF TABLES

Table 1: Competition Matrix of Different LIB Recycling Technologies	8
Table 2: Batteries Sorted in This Study.....	9
Table 3: Competition Matrix of Direct Recycling Technologies	13
Table 4: Graphite ICP-MS Data Tracking Metal Impurity Content.....	18
Table 5: NCM111 and NCM622 ICP-MS Data Showing Relithiation in Recycled Cathode	24
Table 6: Benefit Projections Comparing Direct Recycling with Pyro and Hydro Processes	37

Executive Summary

Lithium-ion batteries are central to California's transition to renewable electricity and zero-emission transportation. This transition is driven by ambitious goals established by Senate Bill 100, which targets 100 percent renewable electricity by 2045, and Executive Order N-79-20, which targets 100 percent zero-emission light-duty vehicle sales by 2035. Achieving these milestones will require extensive deployment of lithium-ion batteries in energy storage systems and plug-in electric vehicles that will create a significant electronic waste stream as these batteries reach the end of their lifecycle. Without sustainable management, the rapid consumption of lithium-ion batteries risks resource shortages and price volatility for critical materials like lithium, cobalt, and nickel — key contributors to battery costs. Recycling and recovery of these valuable materials, which can constitute 45–60 percent of battery manufacturing costs, are essential to lowering production expenses and reducing the lifecycle environmental hazards posed by improper disposal. This underscores the urgent need for effective recycling, reuse, and remanufacturing strategies to recover resources and mitigate the ecological impact of battery waste.

Several advanced technologies have been developed for lithium-ion battery recycling, with most research focusing on cathode materials due to their higher value and complex compositions. The three primary approaches — pyrometallurgical, hydrometallurgical, and direct recycling — each have distinct advantages and limitations. Pyrometallurgical recycling relies on high-temperature smelting, which is energy-intensive and generates significant pollutants. Hydrometallurgical recycling employs strong acids and oxidants to extract metals, necessitating extensive treatment to address environmental and safety concerns. In contrast, direct recycling emphasizes the physical separation and regeneration of cathode and anode materials, preserving their intrinsic value while minimizing environmental impact.

Project Purpose and Approach

In this project, the University of California, San Diego (UCSD) collaborated with Smartville Inc. and American Lithium Energy Corporation (ALE) to develop and scale efficient, high-value direct recycling pathways for lithium-ion batteries. UCSD contributed extensive expertise in regeneration techniques, achieving materials with performance comparable to pristine counterparts and advancing scalable, intensified processes. Smartville leveraged its expertise in battery analysis, de-energization, and second-life battery refurbishment, while ALE focused on assessing the viability of recycled materials by testing their performance in pouch cells with commercial loading under real-world operational conditions. This partnership successfully optimized separation and regeneration workflows, ensuring high efficiency, enhanced safety, and strong commercial potential; details are outlined in subsequent sections.

Significant progress was made in advancing direct recycling despite the challenges posed by the complex composition and structure of lithium-ion batteries. Laboratory-scale operations included battery sorting, materials separation, and hydrothermal regeneration of cathodes and graphite anodes, producing regenerated materials with an electrochemical performance

matching that of new materials. Laboratory processes using 100-gram batches were validated through structure and composition analysis and rigorous cell testing. Scaling efforts increased batch sizes to 5 kilograms under industry-relevant conditions, employing flotation cells, reactors, and furnaces and advancing the technology to demonstration in a real-world operational environment. Closed-loop modeling highlighted the environmental and economic benefits of direct recycling over traditional pyrometallurgical and hydrometallurgical methods. These advancements provide a foundation for developing a 100-kilogram-scale pilot facility and advancing commercial adoption. With the anticipated growth in battery volumes, California is well-positioned to establish in-state recycling facilities, reduce waste, enhance resource recovery, and support the transition to electrified transportation and renewable energy.

Key Results

The project successfully met all key performance metrics and objectives, achieving significant advancements in the direct recycling of lithium-ion batteries. A robust methodology was developed for the deactivation, dismantling, and separation of spent battery components that emphasized safety and energy efficiency. The process enabled the recovery of over 90 percent of cathode and anode active materials, with a froth flotation technique yielding a cathode black mass of over 97-percent purity, suitable for direct regeneration. The direct regeneration process, developed and patented as Purification and Regeneration Integrated Materials Engineering (PRIME), further improved efficiency by integrating impurity removal into the workflow, producing high-quality cathode materials that matched the performance of commercial counterparts.

Electrochemical testing at UCSD validated the recovered lithium cobalt oxide cathode material, which achieved a first-cycle discharge capacity of 143 milliampere-hours/gram (mAh/g) mass and a Coulombic efficiency of 93.46 percent, comparable to virgin materials. Similar results were observed for other cathode chemistries, including NCM111 and NCM622, demonstrating complete regeneration with strong impurity tolerance. NCM111 and NCM622 are types of lithium-ion batteries that use a cathode made of nickel, cobalt, and manganese. The numbers refer to the specific ratio of nickel, cobalt, and manganese. A pilot-scale 5-kilogram regeneration process further verified scalability. Subsequent fabrication and testing of 1-ampere-hour (Ah) pouch cells at ALE's facility demonstrated practical applicability, with the recovered NCM111 delivering a capacity of 0.92 Ah, an initial Coulombic efficiency of 99.2 percent, and 98-percent capacity retention after 75 cycles, confirming the high performance and reliability of the regenerated materials.

Additionally, a comparative analysis of traditional pyrometallurgical and hydrometallurgical recycling methods highlighted the economic and environmental advantages of the PRIME process. By eliminating toxic solvents like N-Methyl-2-pyrrolidone and dimethyl carbonate, the process reduces material and operational costs while minimizing greenhouse gas emissions, making it both sustainable and commercially viable. The PRIME process results in 55 percent lower greenhouse gas emissions compared to conventional pollutive recycling methods, primarily due to its minimal chemical input. Additionally, the estimated profit is 40 percent higher than that of the most remunerative pollutive recycling routes. This positions the PRIME

process as an ideal choice for an environmentally aligned recycling solution. The collaboration among UCSD, Smartville Inc., and ALE showcased an end-to-end recycling solution that addresses economic, environmental, and industrial challenges, paving the way for broader adoption and larger-scale implementation.

Knowledge Transfer and Next Steps

As part of this project's effort to accelerate the commercialization of sustainable lithium-ion battery recycling, UCSD successfully transferred its patented PRIME process to ExPost Technology (ExPost), a UCSD spin-off company based in San Diego. This transfer marks a major milestone in bridging academic innovation with industrial implementation. Through detailed technical documentation, direct training, and joint validation efforts, UCSD provided ExPost with the process know-how necessary to reproduce and scale the technology independently.

ExPost has since commissioned a suite of pre-pilot equipment to implement the PRIME process at kilogram-to-tens-of-kilograms scale. This includes high-throughput shredding, sieving, magnetic separation, and chemical regeneration systems to return lithium to the cathode material of spent batteries, culminating in a 100-liter reactor capable of batch-processing up to 30 kilograms of cathode material. Additional post-processing steps, such as vacuum drying, high-speed mixing, and atmosphere-controlled sintering were also developed to ensure uniform lithium incorporation and restored electrochemical performance.

This successful transfer of knowledge and technology from UCSD to ExPost lays the groundwork for establishing California-based industrial-scale direct recycling operations. The outcomes of this collaboration demonstrate a replicable model for moving university-led clean energy innovations into commercial practice.

CHAPTER 1:

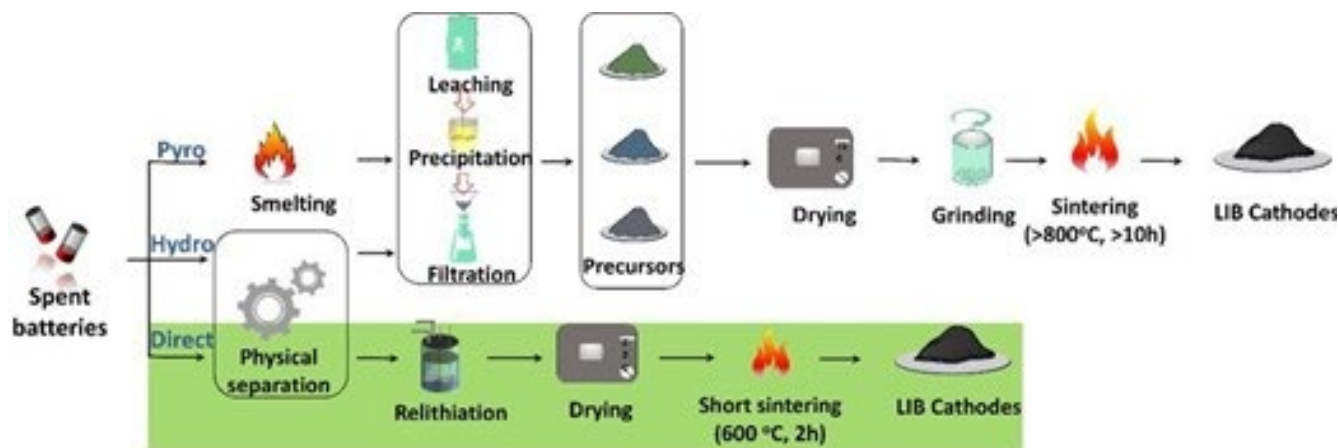
Introduction

Lithium-ion batteries (LIBs) are poised to play a pivotal role in California's transition to zero-emission electricity and transportation in the coming decades. Senate Bill 100 (De León, 2018) establishes the goal of achieving 100 percent renewable and zero-carbon electricity for retail electric sales by 2045. Preliminary analyses suggest that meeting this target could require cumulative additions of 50 gigawatts (GW) in grid-connected battery storage by 2045, much of which is expected to rely on LIBs. Similarly, electrification of transportation will demand even greater deployment of LIBs, particularly in plug-in electric vehicles. Executive Order N-79-20 mandates that all passenger vehicles sold after 2035 must be zero-emission, with medium- and heavy-duty commercial vehicles transitioning to zero-emission where feasible by 2045. This combined growth in energy storage systems and electric vehicle (EV) batteries will generate a significant electronic waste stream as these batteries reach the end of their lifecycle, necessitating sustainable management strategies.

Without intervention, the massive consumption of LIBs could lead to resource shortages and price surges for critical materials such as lithium and transition metals like cobalt, nickel, and manganese. Economically recovering these valuable metals — estimated at approximately (~) \$24–25/kilogram (kg) for cobalt, ~ \$15–20/kg for nickel, and ~ \$8-10/kg for lithium (London Metal Exchange, 2024) — can substantially lower the overall cost of LIB manufacturing, where cathode and anode materials account for 30–40 percent and 15–20 percent of costs, respectively (Gupta et al., 2024). Conversely, the improper disposal of used LIBs exacerbates environmental risks, since flammable and toxic wastes such as organic solvents and heavy metals can cause severe pollution if not carefully treated (Chen et al., 2016). To address these challenges, it is imperative to recycle, reuse, and remanufacture LIBs, enabling both the recovery of valuable materials and the mitigation of environmental impacts.

This project focused on developing and scaling advanced direct regeneration technologies for spent LIBs, aiming to recover valuable materials while minimizing environmental hazards. Conventional pyrometallurgical (pyro) and hydrometallurgical (hydro) recycling methods focus on recovering elemental materials and precursors for cathode production. In contrast, the direct recycling approach, shown in Figure 1, aims to directly recover the higher-value active materials of LIBs while emphasizing safety, environmental sustainability, and cost-effectiveness.

Figure 1: Simplified Schematic Comparison for Recycling Processes



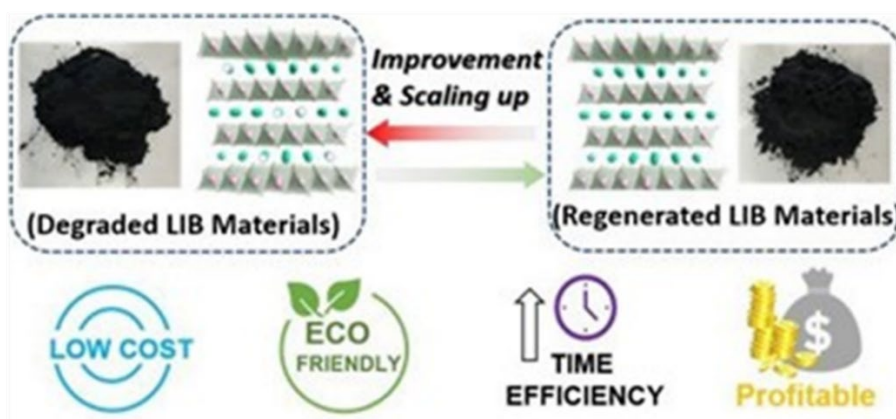
Source: UCSD

Specifically, the project sought to optimize de-energization processes for retired LIBs with various chemistries, to improve cathode and anode separation processes, and to develop robust, universal, energy-efficient, and cost-effective direct recycling methods for diverse LIB materials. Additionally, it aimed to fabricate new LIB cells using regenerated materials and evaluate their performance relative to virgin materials, while conducting life-cycle analyses to assess the energy, economic, and environmental impacts of direct recycling. The ultimate goal was to establish a commercially viable pathway for recycling spent LIBs into high-quality materials suitable for new cell manufacturing, thereby reducing production costs, lowering greenhouse gas emissions, and accelerating the adoption of advanced battery technologies (Figure 2).

The complicated composition and structure in LIBs make regeneration of valuable active materials difficult. The University of California, San Diego (UCSD) team demonstrated significant progress in laboratory-scale operations, including battery sorting, materials separation, and hydrothermal direct regeneration of various LIB cathodes and graphite anodes. Using 100-gram (g) batch processes, the team successfully regenerated materials with an electrochemical performance on a par with pristine commercial materials. This was validated through structure and composition analyses as well as testing in coin cells (3–4 mAh), achieving Technology Readiness Level (TRL) 4¹ or process validation in a laboratory environment. Closed-loop modeling of energy consumption, environmental impact, and costs further underscored the advantages of the direct recycling approach over traditional pyrometallurgical and hydrometallurgical processes.

¹ Technology Readiness Level (TRL) 4 indicates validation of a technology in a laboratory environment. At this stage, basic technological components are integrated to establish that they work together.

Figure 2: Illustration of Technology and Its Advantages



Source: UCSD

To scale up operations from 100-g to 5-kg under industry-relevant conditions, this project involved sorting and de-energizing LIB cells with over 1,000 ampere-hours (Ah) total capacity, equivalent to hundreds of 18,650 cylindrical cells (for example, Tesla cells) or 20–30 pouch cells (for example, GM Volt cells) per batch. Materials separation was conducted using froth flotation and manual disassembly, which efficiently separated cathode and anode components with high purity. Scaling to a 5-kg regeneration process required increasing the flotation cell volume and adopting a 20L (liter) reactor, a 20-fold increase over the conventional lab scale setup. The final step involved short annealing in a furnace. Completion of these tasks, along with data collection and process modeling, advanced the technology to TRL 6,² i.e., the process was demonstrated in a real-world operational environment.

The insights gained from this project provide a strong foundation for developing a larger pilot facility capable of operating at a 100-kg scale under industry conditions, paving the way for commercial adoption. Though no large-scale lithium battery recycling facilities are located in California today, this fact will change as available used battery volumes increase and as the logistics costs of transporting batteries out of state become too burdensome and expensive. As a result, the assumption can be made that close to 100 percent of used batteries in California will eventually be recycled in-state once volumes justify the required investments in recycling facilities and once supply-chain logistics are established. Battery manufacturers and automakers could leverage this recycling pathway to enhance efficiency, minimize waste, and support the broader electrification of the automotive industry.

² TRL 6 refers to a system or prototype demonstrated in a relevant environment. It indicates that the technology has been tested at near-operational scale under conditions similar to real-world use.

CHAPTER 2:

Project Approach

Many technologies have been explored for LIB recycling. Due to the higher value of cathode materials than anode materials (graphite), as well as their complex compositions, most of the research and development have been focused on cathodes. The three general approaches for LIB recycling (Table 1) are: pyro, hydro, and direct recycling processes. The pyro (smelting) process uses a high-temperature furnace to destroy battery components and generate metal (or alloy) ingots. This process is energy intensive and generates air pollutants (Zhang and Xu, 2016). The hydro process uses strong acids (for example, sulfuric acid [H_2SO_4]) and/or oxidative reagents (e.g., hydrogen peroxide [H_2O_2]) to extract critical metals from cathode active material into solution, followed by complicated refining processes to recover metal salts. The extensive use of corrosive and toxic chemicals requires special treatment to mitigate the pollution issue (Tuncuk et al., 2012). Pyro and hydro processes result in elemental products such as cobalt sulfate (CoSO_4), nickel sulfate (NiSO_4) and lithium carbonate (Li_2CO_3).

In comparison, the direct recycling process focuses on physical separation of electrode materials, including electrolytes, metal current collectors, plastic separators, and cathode and anode active materials. The latter two components, which represent the majority of the battery value, are treated using regeneration methods to restore their electrochemical properties without going through a complicated destructive process. Direct recycling is one of the most efficient technologies to reintroduce the value-concentrated cathode material into the supply chain with the lowest environmental impact.

Ideally, products obtained from direct recycling can be readily used by battery manufacturers, which can lower the cost of cell production. The Sustainable Power and Energy Center (SPEC) at UCSD collaborated with Smartville Inc. and American Lithium Energy Corporation (ALE) to advance and fully develop a high-value, efficient, and scalable direct recycling pathway for LIBs across a diverse range of cell chemistries. UCSD contributed its extensive expertise from previous research projects funded by the California Energy Commission (CEC), U.S. federal agencies, and private companies. UCSD also collaborated with the U.S. Department of Energy's ReCell Center, automotive manufacturers (General Motors and Honda), and the National Science Foundation (NSF) to design and enhance solution-based direct regeneration methods for LIB cathode materials and graphite anodes. These efforts demonstrated regenerated materials with performance on a par with pristine materials. Additionally, UCSD's team achieved significant process intensification, paving the way for scalable operations while maintaining efficiency and safety.

Table 1: Competition Matrix of Different LIB Recycling Technologies

Comparable Attribute	Pyrometallurgical Recycling	Hydrometallurgy Recycling	Direct Regeneration
Energy consumption	high	moderate	low
Chemical usage	high	high	low
Process complication	high	high	low-moderate
CO ₂ footprint	high	high	low
Product value	moderate	moderate	high
Profit	low	low-moderate	high

Source: UCSD

Smartville Inc. applied its experience in cost-effective LIB analysis, de-energization, and the refurbishment of second-life EV batteries for commercial and utility-scale energy storage, supported by the CEC (EPC-19-038), the U.S. Department of Energy (DOE) (Battery Recycling Prize), and other programs. ALE, with over a decade of experience in LIB research, development, and manufacturing for military and commercial applications, focused on evaluating the viability of recycled materials by testing their performance in cells under operational conditions.

Together, the partnership leveraged complementary capabilities to advance key operational processes, including battery and material separation, high-value material regeneration, and comprehensive process modeling to assess the energy, economic, and environmental impacts across the entire LIB life cycle. By building on their understanding of solution chemistry, interface engineering, and solid-state electrochemistry, the team members tailored each operational step to achieve optimized temperatures, short processing times, high product quality, and minimal waste, ensuring the technology's appeal for commercial adoption. Individual accomplishments are discussed in the subchapters.

Battery Acquisition, Diagnosis, Sorting, and De-energization

The primary goal of this task was to prevent battery destruction, hazardous degassing, corrosion, and excessive heat generation during the de-energization process while avoiding the creation of hazardous waste during storage or transportation. This project innovated battery recycling logistics by (1) enabling quick health assessment and initial discharging before slow draining, thus improving efficiency, and (2) producing fully inert batteries for safe sorting and batch transfers, thereby enhancing recycling efficiency and safety.

For this project, 25 pounds of batteries with various chemistries, types, ages, and usage histories were acquired (Figure 3), including nickel manganese cobalt (NMC), lithium iron phosphate (LFP), lithium manganese oxide (LMO), and nickel cobalt aluminum (NCA) batteries for diagnosis, sorting, and de-energization. These batteries were sourced from diverse industries, such as mobility, stationary storage, electronics, and niche applications, to ensure a comprehensive evaluation of the recycling process. The project team carefully divided the 25

pounds of batteries among different cell types, as outlined in Table 2, to represent a wide range of compositions and conditions. This diverse selection facilitated the development and optimization of processes for battery sorting and recycling, ensuring scalability and applicability across multiple battery technologies and industries.

Figure 3: Battery Modules Collected From Various Sources



Source: Smartville

Table 2: Batteries Sorted in This Study

Cell type	Chemistries	Manufacturer	Number of cells	Module disassembled	Cell weight
Cylindrical	LFP	A123	100	2	76 g
Pouch	LMO	Nissan	8	2	919 g
Pouch	NMC	LG Chem	16	1	965 g
Cylindrical	NCA	Panasonic	330	1	47 g

Source: Smartville

Initial diagnosis was conducted based on a battery health assessment. A comprehensive physical inspection was conducted to assess the battery's external condition. This included checking for visible damage such as cracks, dents, or swelling. Additionally, an insulation resistance test was performed to evaluate the integrity of the battery's insulation. Later sorting was done based on state-of-health (SOH) metrics, including capacity and impedance testing, and cathode chemistry.

To assess the SOH of LIBs, the project team employed a combination of two aging variables: internal resistance and remaining capacity.

- **Internal Resistance:** DCIR, or Direct Current Internal Resistance, measures the resistance within a battery that opposes the flow of electrical current. A higher DCIR indicates increased internal resistance, leading to reduced performance.
- **Remaining Capacity:** The total capacity delivered during the discharge test provides insight into the battery's remaining usable capacity. Specifically, the pulse discharge test is used to measure two key parameters (See Appendix A for relevant protocols and calculations).

Smartville Inc. developed an advanced system incorporating state-of-charge (SOC) and SOH estimation, as well as SOC and life balancing algorithms, which were implemented in MATLAB. These algorithms were designed to sort and recondition batteries, improving SOH uniformity, performance, and reliability for subsequent repurposing or recycling applications. The SOC estimation algorithm was validated using acquired battery cell data, while the SOH estimation algorithm was fine-tuned for the specific characteristics of these cells.

The objective of de-energization is to reduce the risk of short-circuiting and self-ignition in spent LIBs. To achieve this, LIBs must be fully discharged to eliminate any residual electrical charge. In this project, Smartville evaluated various resistive shunting methods for safely discharging the batteries. Based on the findings, it was concluded that the safest and most non-intrusive method for discharging both modules and individual cells involves discharging to a specific voltage limit, followed by slow discharging across resistors. To ensure a safe and controlled de-energization process, a two-step discharging protocol was implemented (see the protocol in Appendix A).

All cells in the project were de-energized by discharging them to below 0.1 volts (V) using resistors to ensure safety during recycling. The task goals were successfully met, with safe and rapid sorting and deactivation of over 200 cells per batch for shredding and separation.

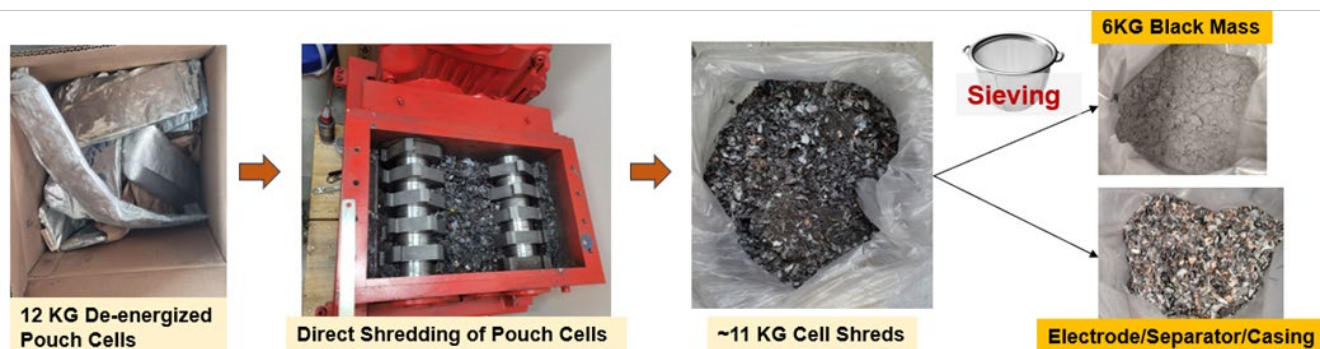
Cathode and Anode Materials Separation

For the extraction of cathode black mass (CBM) on a gram scale in a standard laboratory setting, batteries are first discharged then manually disassembled to obtain cathode strips. The electrolyte residue is then dried off, and CBM is scraped off the strips using a blade. However, scaling this process to the kilogram level renders manual scraping impractical. Therefore, active material extraction by comminution at the cell level was implemented, followed by sieving to assess the yield and quality of the feedstock. For the regeneration trials, post-sieving active material separation was necessary to obtain the CBM from the black mass. However, the primary goal of this experiment was to set up the appropriate equipment for the preprocessing extraction of kilogram-level active material powder at UCSD.

Over 12 kg of large pouch cells (3.8" x 19.5") were received from a vendor (A) for the black mass extraction trial (Figure 4). The pouch cells were perforated and left in a fume hood to allow the initial release of the electrolyte. After drying, the 12 kg of cells were shredded and then dried again to remove any remaining electrolyte, thereby preventing blockage during the

sieving process. Sieving was conducted using an 80 mesh (177 microns) in a sealed vibratory sieve shaker placed within a fume hood for safety. The operation yielded approximately 6.62 kg of black mass, corresponding to a yield of about 53.54 percent relative to the initial cell weight. Excluding the weight of other components, such as electrode shreds, separators, and casings, the overall yield of the process after shredding and sieving was approximately 90.87 percent, with 9.13 percent of the weight lost, primarily as dried electrolyte and other process losses.

Figure 4: Kilogram Level Black Mass Recovery for Scaling up Regeneration

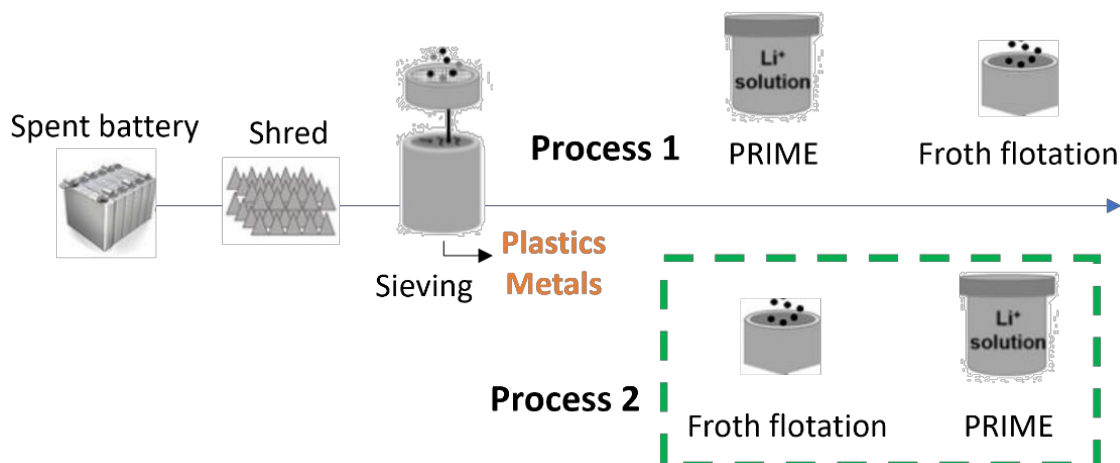


Source: UCSD

After spent LIBs are shredded and sieved, fine powders (10–50 micrometers [μm]), primarily comprising anode and cathode materials (collectively termed “black mass”), can be obtained through physical separation methods. In this project, an efficient and environmentally friendly froth flotation method was employed to separate cathode and anode particles. Froth flotation operates on the principle of segregating materials with different hydrophobicity (physical property of a substance to repel water) in an aqueous medium. The hydrophobicity difference between LIB cathode materials and graphite allows for high-efficiency separation. Due to its oxide surface, layered oxide-based cathode active material (CAM) typically exhibits good hydrophilicity (the property of being attracted to water). However, the presence of a hydrophobic binder and carbon additives on the CAM surface can reduce separation efficiency.

Some research groups have utilized pre-burning to decompose binders at temperatures exceeding 572°F (300°C), which improves separation efficiency. However, this approach introduces residual fluorine on the CAM surface, compromising the performance of the recovered material. This project used a different approach, where froth flotation was directly evaluated on black mass samples without preheating. Additionally, the purification and regeneration integrated materials engineering (PRIME) process (discussed in the section titled “Improve Efficiencies of Direct Recycling Processes”), revealed that the binder could be degraded during hydrothermal treatment, reducing CAM hydrophobicity. This improved the separation of CAMs from graphite materials post-PRIME. Figure 5 outlines two workflows for separating layered oxide-based CAMs after shredding.

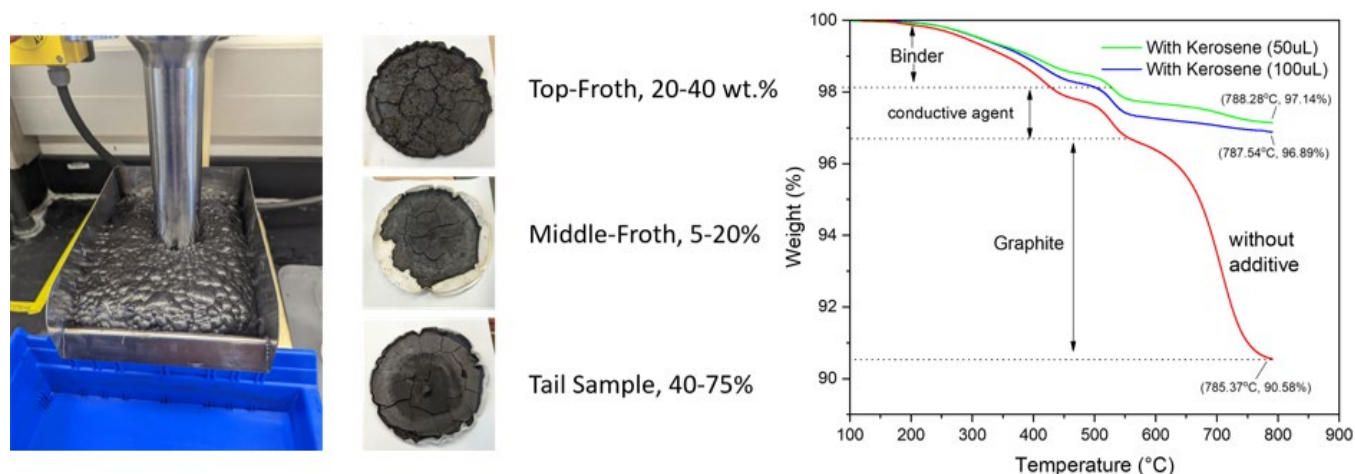
Figure 5: Separation Workflow From Shredding to Froth Flotation



Source: UCSD

Results from Process 2 were more promising and were selected for further development. For the optimization, the UCSD team started with the solid-to-liquid (S/L) ratio, as the pulp density drastically changed the particle dynamic behavior during the froth flotation process. Three different ratios were selected for the initial assessment. Besides the S/L ratio, additives optimization was conducted. Based on the literature investigation, the UCSD team identified kerosene as an effective additive for CAMs and graphite separation during froth flotation. Kerosene is a commonly used non-ionic collector for froth flotation, specifically in the coal industry. The process involves the selective adsorption of oils by minerals. The kerosene amount was investigated when controlling all other parameters and it was found that a small amount of kerosene, specifically, 50 microliters per 2.5 liters of water would drastically improve the separation efficiency of CAMs from graphite particles. (The complete protocol is contained in Appendix A.) In the recovered product after froth flotation separation, the sample purity was found to be more than 97 wt. percent as indicated by thermogravimetric analysis (Figure 6), which implied that the sample was pure enough for the subsequent regeneration process.

Figure 6: Froth Flotation Separation and Purity Analysis of the Product



Source: UCSD

Improve Efficiencies of Direct Recycling Processes

Current direct recycling processes are primarily based on recovering the composition and structure of degraded active materials and then performing physical/chemical treatment to regenerate active materials. However, accurate determination of the composition and structure of degraded LIBs from different resources is challenging, which makes viability questionable. Table 3 compares the features of different direct recycling approaches.

Table 3: Competition Matrix of Direct Recycling Technologies

Comp. Attribute	Solid-state	Electrochemical	Ionothermal	Hydrothermal
Energy consumption	high	moderate	low	low
Product quality	low	low	low	high
Scalability	high	low	low	high
Major limitation	low product quality	not scalable	not scalable	pressure needed
*Process cost (\$)	10	N/A	20	7.5
**Process profit (\$)	10	N/A	0	12.5

* Cost is based on 1 kg of NMC111 cathode materials — EverBatt model

** Assumes \$20/kg for NMC111.

Source: UCSD

Hydrothermal regeneration takes precedence as the most practical and environmentally benign solution, offering a low-cost, water-based treatment method. Many studies have demonstrated hydrothermal relithiation³ as an effective direct regeneration approach, successfully recycling cathodes like spent lithium cobalt oxide (LCO) (Shi, Chen, and Chen, 2018; S. Sloop et al., 2020), $\text{LiNi}_{0.33}\text{Co}_{0.33}\text{Mn}_{0.33}\text{O}_2$ (NCM111) (Shi, Chen, Liu, et al., 2018), $\text{LiNi}_{0.5}\text{Co}_{0.2}\text{Mn}_{0.3}\text{O}_2$ (NCM523) (Shi et al., 2019), $\text{LiNi}_{0.6}\text{Co}_{0.2}\text{Mn}_{0.2}\text{O}_2$ (NCM622) (S. E. Sloop et al., 2019), and LiFePO_4 (Xu et al., 2020) cathodes back to a pristine state in terms of composition, structure and performance. The process uses a minimal amount of chemicals and does not generate wastewater. Additionally, the hydrothermal process was found to be self-saturating, meaning it could restore the stoichiometry of active material degraded to varying degrees back to the same level, thereby eliminating the need for extensive analysis of feedstock from different sources (Gupta et al., 2023).

Cathode Recovery

Ensuring the compositional and structural purity of the CAM is a significant challenge in recycling. The cathodes from end-of-life (EoL) batteries often contain various impurities, such as the electrolyte salt (usually LiPF_6), conductive carbon (usually Super P65/P45), aluminum fragments, and binder (usually polyvinylidene fluoride [PVDF]) (Gupta et al., 2024). Conversely, graphite, which serves as the typical anode active material (AAM), exhibits exceptional stability compared to CAMs. Anodes from EoL batteries have similar impurities as

³ Relithiation is the process of restoring lost lithium ions into a lithium-ion battery cathode material to recover its original electrochemical performance.

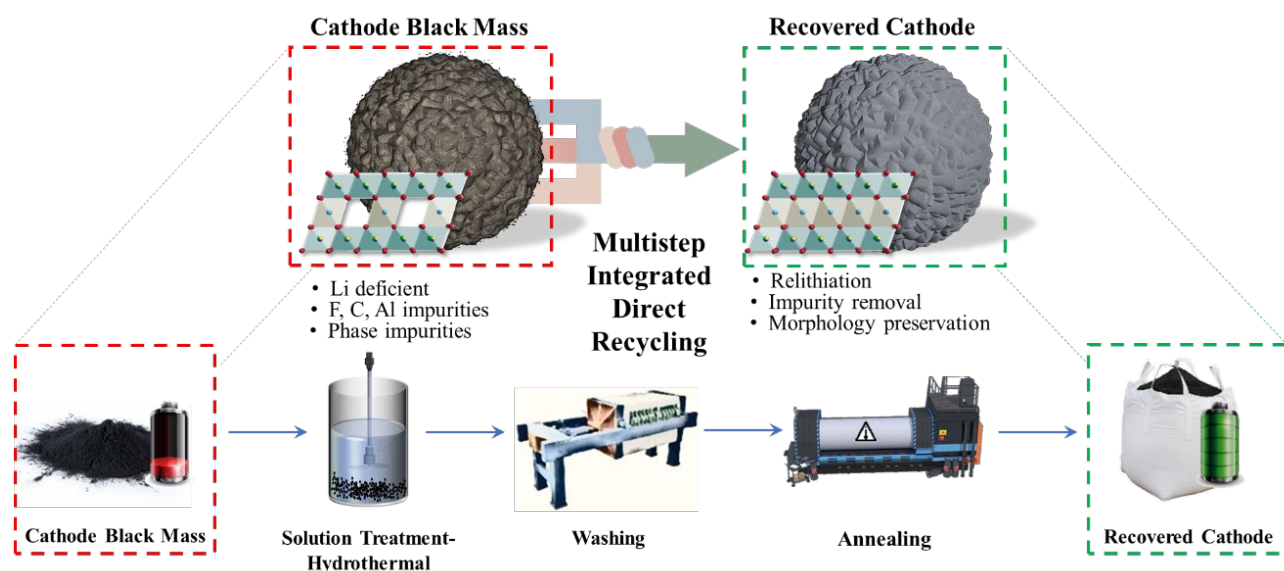
cathodes, with the binder commonly replaced by water-soluble alternatives (typically carboxymethyl cellulose or styrene butadiene rubber) and aluminum substituted by copper shreds. The powder obtained from cathodes and anodes with impurities is called “cathode black mass” (CBM) and “anode black mass” (ABM), respectively. The impurity removal is concisely called purification (Gupta et al., 2023). In CBM, fluorine-based impurities pose the most significant challenge during direct recycling.

The difficulty arises from the toxicity and stability of the decomposed products, making their removal during the recycling processes challenging. PVDF binder (typically 3-4 wt. percent) and lithium hexafluorophosphate (LiPF_6) electrolyte salt (typically 1.3 wt. percent) are the two main sources of fluorine present in the cell (Murdock et al., 2021). PVDF is typically challenging to remove due to its resistance to most chemicals, high abrasion resistance, thermal stability up to 662°F (350°C), and solubility only in specific organic solvents (Saxena and Shukla, 2021).

During this project, a scalable process, called PRIME, was devised that combines purification and relithiation techniques for CBM, ensuring high-quality output. The direct recycling process purifies CBM by integrating PVDF decomposition within the relithiation process. Design of the process is shown in Figure 7. The three main steps include:

- 1) Solution treatment such as a hydrothermal relithiation process with PVDF decomposition and electrolyte salt removal (operation time, 4-6 hours).
- 2) Washing and drying, for extra alkaline solution, conductive carbon, degraded PVDF, and other impurity removal (operation time, 3-6 hours).
- 3) Annealing, for crystal structure repair and remnant carbon removal (operation time, 4-8 hours).

Figure 7: Schematic of the Process Indicating Scalable Regeneration of CAM by Multistep Integrated Direct Recycling

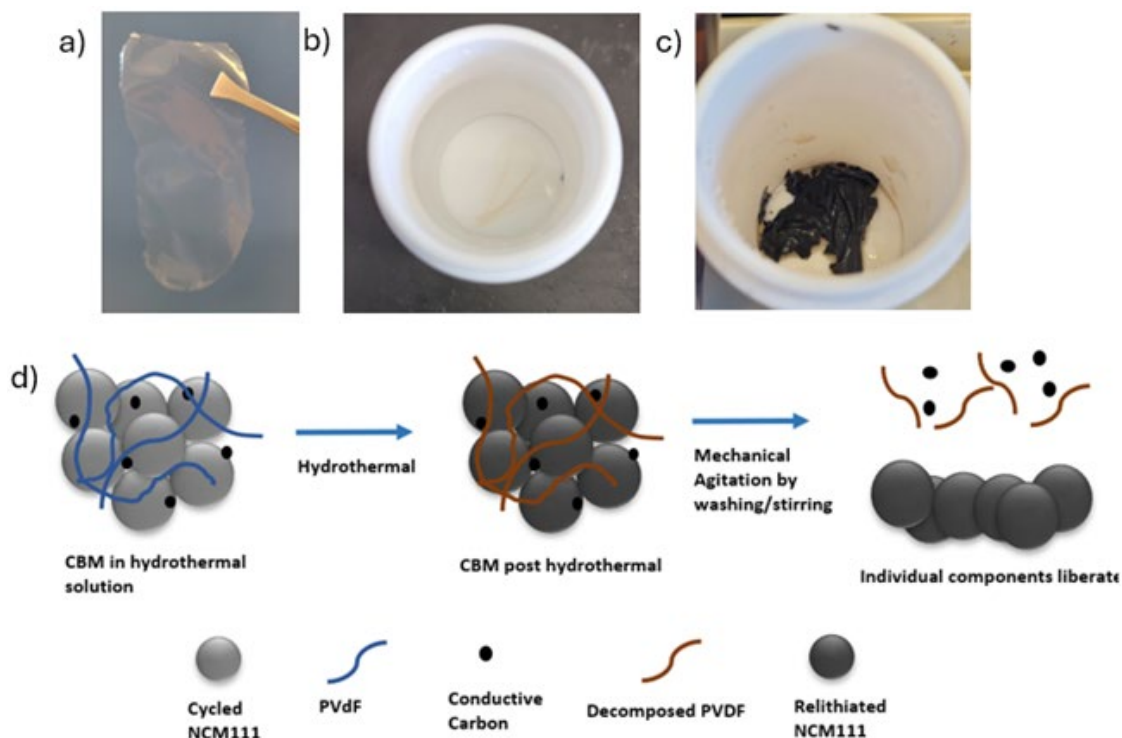


Source: UCSD

Notably, the PRIME process avoids the use of organic solvents and successfully regenerates CBM to its original state.

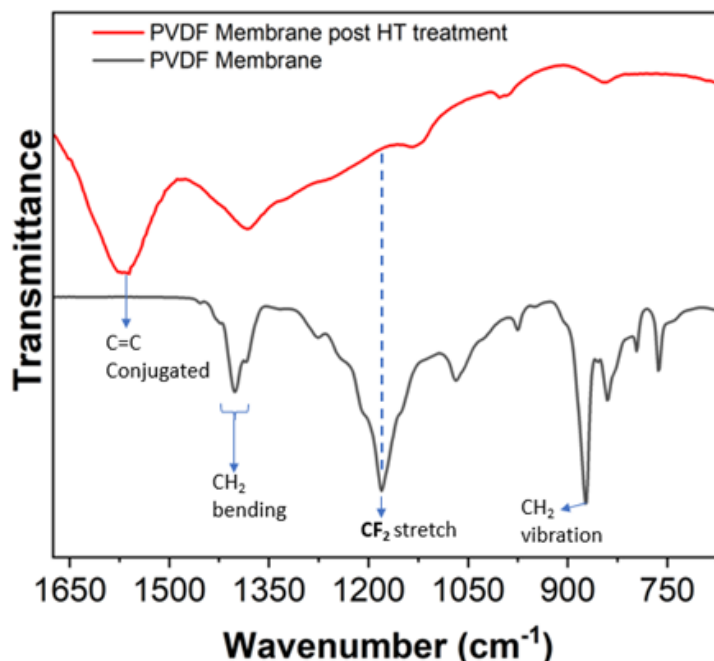
The investigation into the successful integration of PVDF binder removal in the hydrothermal step was also carried out. The prior research showcased the effectiveness of high-temperature relithiation in an alkaline environment as a crucial process for rejuvenating cathode materials (Shi, Chen, and Chen, 2018; Shi, Chen, Liu, et al., 2018). Therefore, by utilizing the hydroxides/alkoxides present in the solution to interact with the PVDF binder, the efficiency of the relithiation conditions can be further enhanced. To elucidate the mechanism of PVDF degradation during the high-temperature process, a pure PVDF membrane (Figure 8a) was fabricated and subjected to high-temperature hydrothermal reaction in an alkaline solution (Figure 8b), employing the same conditions used for cathode relithiation. Following the reaction, the initially transparent PVDF membrane transformed into a black, fragile membrane (Figure 8c). Fourier transform infrared spectra (FTIR) of the dried decomposed membrane (Figure 9) revealed the disappearance of characteristic fluorocarbon peaks (1,000–1,250 inverse centimeters [cm^{-1}]) observed in the original PVDF membrane, while a broad peak for double-bonded carbon appeared at $1,600\text{ cm}^{-1}$. This result aligns with earlier studies suggesting that PVDF membranes undergo defluorination reactions under alkaline environments (Ross et al., 2000). Hence, two key findings were confirmed: firstly, PVDF can undergo defluorination/decomposition under the same conditions utilized for cathode relithiation and, secondly, the resulting decomposed product loses its mechanical integrity to the extent that it can be broken by agitation during washing. The overall mechanistic understanding is illustrated in Figure 8d.

Figure 8: Illustration of PVDF Decomposition Mechanism During PRIME Process



Source: UCSD

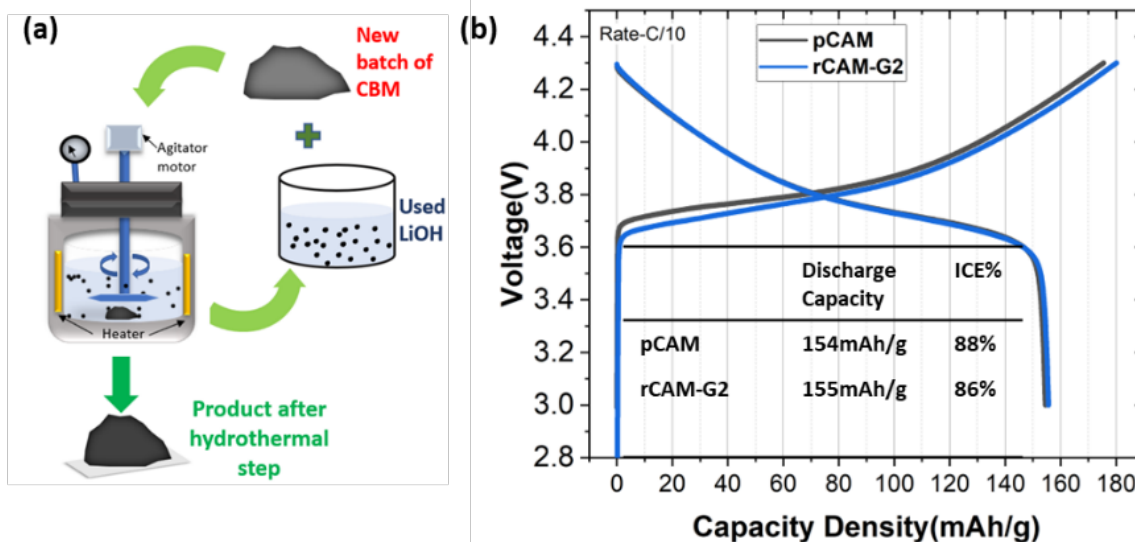
Figure 9: FTIR Spectroscopy Analysis of Decomposed Binder (PVDF)



Source: UCSD

Starting with CBM containing approximately 10 percent carbon and PVDF impurities, the PRIME process effectively removed over 99 percent of impurities, demonstrating a tolerance for up to 10 percent reductive impurities. Leveraging established hydrothermal and sintering technologies commonly used in cathode synthesis; the method is safe, straightforward, and scalable to industry-level applications. Furthermore, the potential to reuse chemical solutions, enhancing overall efficiency (Figure 10a-b), was demonstrated.

Figure 10: (a) Solution Reuse Strategy Schematic and (b) Electrochemical Performance of the Recycled Product - Generation 2 (rCAM-G2)

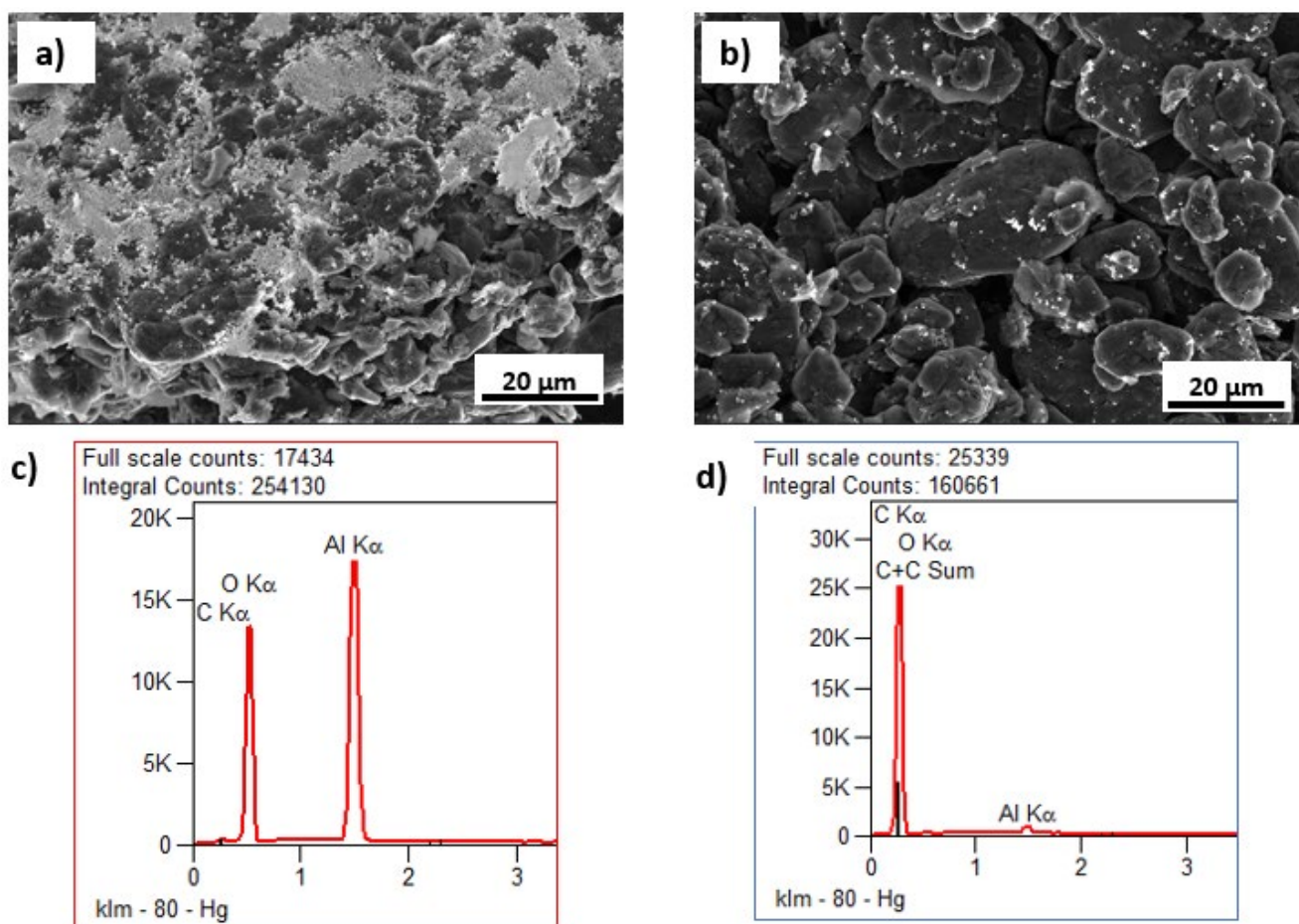


Source: UCSD

Anode Recovery

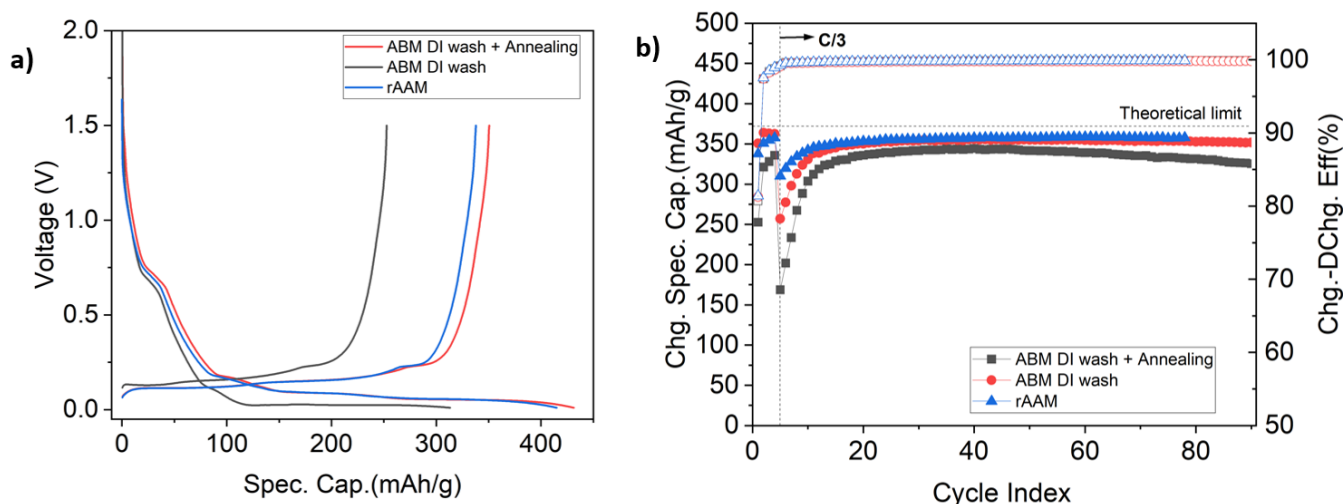
Copper (Cu) is usually the most challenging impurity to remove in ABM because it is relatively stable in comparison to aluminum in CBM. Additionally, irreversible lithium (Li) trapped in the graphite obtained from EoL cells is also treated as an impurity (Markey et al., 2020). The Li amount in bulk graphite may vary based on feedstock. In preliminary analysis of various feedstocks acquired from diverse cell chemistries, the potential for other impurities originating from internal cell components to contaminate the ABM was further noted, such as aluminum oxide (Al_2O_3) from the cathode/separator or transition metals from cathodes. These additional impurities made certain graphite feedstocks more challenging than others. Nevertheless, using a method patented by UCSD, Recycled Anode Active Material (rAAM), the project team successfully purified and upcycled ABM from various feedstocks, achieving greater than (>) 99.5 percent purity and >95 percent discharge capacity recovery (Figure 11 and Figure 12).

**Figure 11: Purified Graphite Product Obtained After Recycling;
Evident From (a-b) SEM and Respective (c-d) EDS* Data**



Key: SEM= scanning electron microscopy, EDS=energy dispersive X-ray spectroscopy.
Source: UCSD

Figure 12: Electrochemical Performance of Recovered Graphite Product (rAAM) Tracked at Each Regeneration Step



Source: UCSD

The capacity degradation in LIBs primarily stems from the loss of Li inventory in the cell, structural changes of the active materials, and impedance growth induced by the formation of a solid-electrolyte interphase on the graphite particle surface (Markey et al., 2020). In addition to the majority of lithium being consumed in the solid-electrolyte interphase, a portion of the lost Li is also irreversibly trapped within the bulk of the graphite particles. Despite this degradation, spent graphite particles typically retain their morphology and bulk structure and therefore require only purification and surface modifications to revert to the high-performance quality graphite (Markey et al., 2020).

For ABM, following purification preprocessing steps, including acid treatment to dissolve copper, transition metals, and impurities, the UCSD-patented boric-acid-based method for graphite upcycling (Figure 13) was employed. The purification procedure involved leaching in a 2 molar sulfuric acid solution for 8 hours, followed by water washing, boric acid washing (1 gram in 2 milliliters [mL] of 5 wt. percent boric acid solution), and 1-hour annealing (1,922°F [1,050°C]) in dinitrogen (N₂). The total process time was close to 12 hours, achieving the high-performance target in accordance with the set metrics. The final graphite product resulted with boron-based surface coating, enhancing electrochemical performance and stability. The results of inductively coupled plasma mass spectroscopy (ICP-MS) confirmed the purification and boron presence in the recovered material (Table 4).

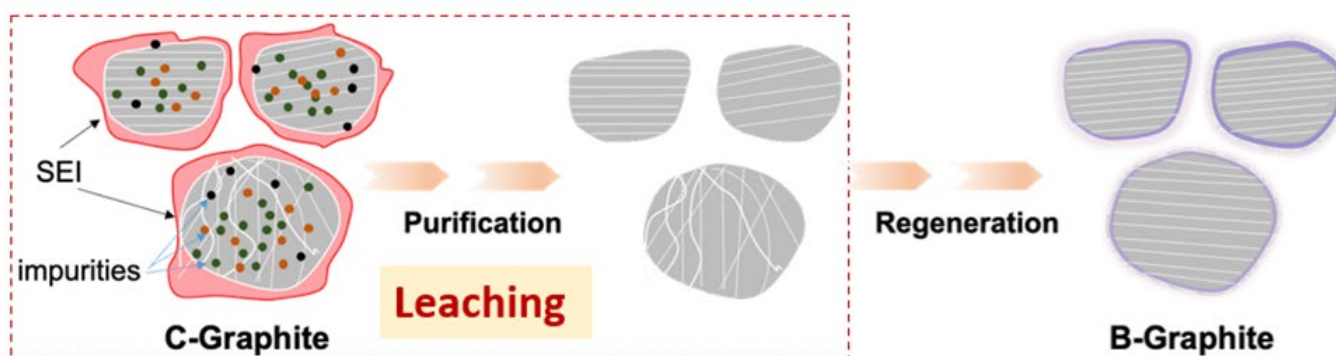
Table 4: Graphite ICP-MS Data Tracking Metal Impurity Content

ABM (cell chemistry)	Li wt%	B wt%	Ni wt%	Mn wt%	Co wt%	Cu wt%	Fe wt%	P wt%
Cycled ABM (NCM111)	0.193	—	0.0137	0.009	0.008	0.931	0.055	0.192
Cycled ABM (NCM622)	0.153	—	0.0161	0.007	0.007	0.713	0.003	0.004

ABM (cell chemistry)	Li wt%	B wt%	Ni wt%	Mn wt%	Co wt%	Cu wt%	Fe wt%	P wt%
Cycled ABM (LFP)	0.052	—	—	—	—	0.052	—	—
rAAM (NCM111)	0.061	0.266	0.024	0.020	0.022	0.755	—	0.100
rAAM (NCM622)	0.110	0.348	0.049	0.006	0.001	0.558	0.263	—
rAAM (LFP)	0.039	0.640	—	—	—	0.052	—	—

Key: LFP=lithium iron phosphate, Li=lithium, B=boron, Ni=nickel, Mn=manganese, Co=cobalt, Cu=copper, Fe=iron, P=phosphorus.
Source: UCSD

Figure 13: Schematic for the ABM Regeneration to Upcycled Graphite

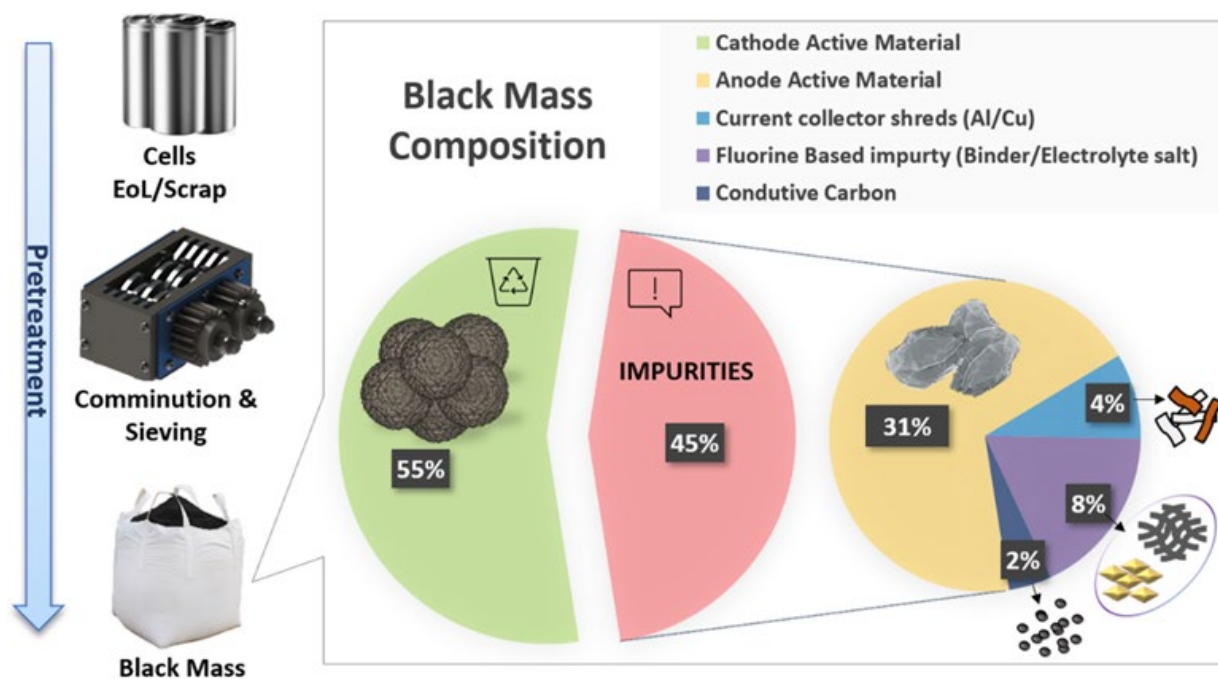


Source: UCSD

Direct Recycling Process Scale-up

The previous section described the developed regeneration process and its tolerance to certain impurity thresholds. Regarding the CAM regeneration, Figure 14 shows that the black mass obtained directly from shredded cells typically contains approximately 45 percent impurities. This impurity level can be reduced to 3 percent under optimal conditions using froth flotation for the LCO single crystal chemistry, as optimized for separation (described in the section titled "Cathode and Anode Materials Separation"), which is different than polycrystalline NCM chemistry. To ensure an appropriate feedstock for NCM direct regeneration and avoid the time-intensive optimization of froth flotation technique, it was deemed necessary to manually disassemble cells to obtain cathode strips to meet the time constraints of the project. These cathode strips were then used as feedstock for the preprocessing operation detailed in the previous section.

Figure 14: Typical Impurities to Deal With After Sieving (<100 microns) to Obtain Feedstock for Direct Recycling



Source: UCSD

The project team received two battery packs from another vendor (B) containing 8 modules and 192 cells, each with a capacity close to 5 Ah. Each module was individually discharged using resistors and then manually disassembled into cells and subsequently into electrode rolls (Figure 15), according to the protocol established the section titled "Battery Acquisition, Diagnosis, Sorting, and De-energization." These rolls were then unwound into their constituent separator, cathode, and anode strips. The cathode strips were subsequently shredded and dried overnight to remove any residual electrolyte. To achieve delamination of the CBM from these cathode electrode shreds, further blending and sieving were performed, ultimately yielding approximately 6.5 kg of CBM product. The yield of CBM relative to the original cells was about 17.1 percent, which approximately aligns with the typical 20–30 percent cathode material-to-cell weight ratio in such cells used for hybrid EV applications (Marshall et al., 2020).

With no graphite impurity, the CBM obtained was an appropriate feedstock for direct regeneration trials, containing less than 10 percent overall impurities, which was within the tolerance limit of the PRIME regeneration process. Therefore, the project team was able to successfully demonstrate an over-5-kg scale recovery of a direct recycling feedstock from a commercial EV pack. It should be acknowledged that the primary time-limiting step in this process was the manual disassembly of cells for cathode strip recovery. Consequently, UCSD is concurrently initiating another project focused on developing automated disassembly methods to enable continuous recycling.

Figure 15: Preprocessing Schematic to Obtain CBM From the Battery Modules Obtained From Spent EV Packs Received From Vendor B



Source: UCSD

Performance Evaluation and Demonstration of Direct Recycling of LIBs

Cathode and anode active material samples from various chemistries of LIBs were recovered, regenerated via direct recycling, and evaluated to be compared with commercial samples. Extensive materials characterization and performance evaluation were integral to the success of this project, enabled by the collaboration between UCSD, Smartville Inc., and ALE.

UCSD's advanced facilities and expertise were pivotal to the project's success. The stoichiometric composition of elements in regenerated particles was analyzed using ICP-MS, a critical tool for assessing lithium deficiency, identifying impurity elements, and confirming comparability to pristine samples. Surface properties, which greatly influence reversible electrochemical performance by side reactions, were characterized via X-ray photoelectron spectroscopy, providing a precise evaluation of impurity levels in the final product after direct recycling. Additional analyses of particle morphology, which dictates the pack density of the electrodes and, therefore, energy density of the cell, were performed using SEM. Direct recycling products need to maintain their morphology without inducing cracks to obtain similar electrode quality as the pristine cathodes. X-ray diffraction (XRD) explains the arrangement of atoms in the crystal and serves as a fundamental check for material chemistry. The quality of the match of the XRD pattern between recovered material by direct recycling and pristine sample defines the success of the regeneration. This comprehensive suite of characterizations offers valuable insights into the processing-structure relationships of regenerated cathode materials, defining their performance after LIB direct recycling.

Electrochemical performance evaluation is a critical step in demonstrating the effectiveness of direct recycling, a process led by UCSD and ALE in this project. Typically, cathode materials in powder form are mixed with a conductive aid and binder to create a slurry, which is then cast onto aluminum foil to produce electrodes. These electrodes are used to fabricate half cells versus lithium metal or full cells versus graphite anodes for a variety of electrochemical tests. Half cells are simpler to assemble and isolate the cathode material as the sole variable, making them ideal for initial performance evaluations of CAM recovered through direct recycling. Once the recycled material demonstrates satisfactory performance across several key metrics, full cells are assembled for further validation.

The first metric assessed from the cells is the voltage-capacity profile, which reveals phase transformations in the cathode material. The initial voltage profile is particularly significant, as it provides an early indication of material quality by reflecting charging and discharging voltages, capacity, and the reversibility of phase transformations, typically quantified through Coulombic efficiency (CE). This initial evaluation is essential for determining whether the recycled material meets performance standards. The kinetics of lithium-ion movement during charging and discharging within the crystal structure are assessed through rate performance tests, which involve charging and discharging at varying current rates. Retention tests at a fixed rate evaluate the material's long-term stability by determining how many cycles it can endure before capacity falls below acceptable levels. These evaluations are necessary to validate the performance of the recycled material against pristine samples. Following successful coin cell testing, the materials are further assessed in pouch cells (1 Ah) under practical operating conditions, testing similar features to confirm scalability and applicability for real-world use.

CHAPTER 3:

Results and Discussion

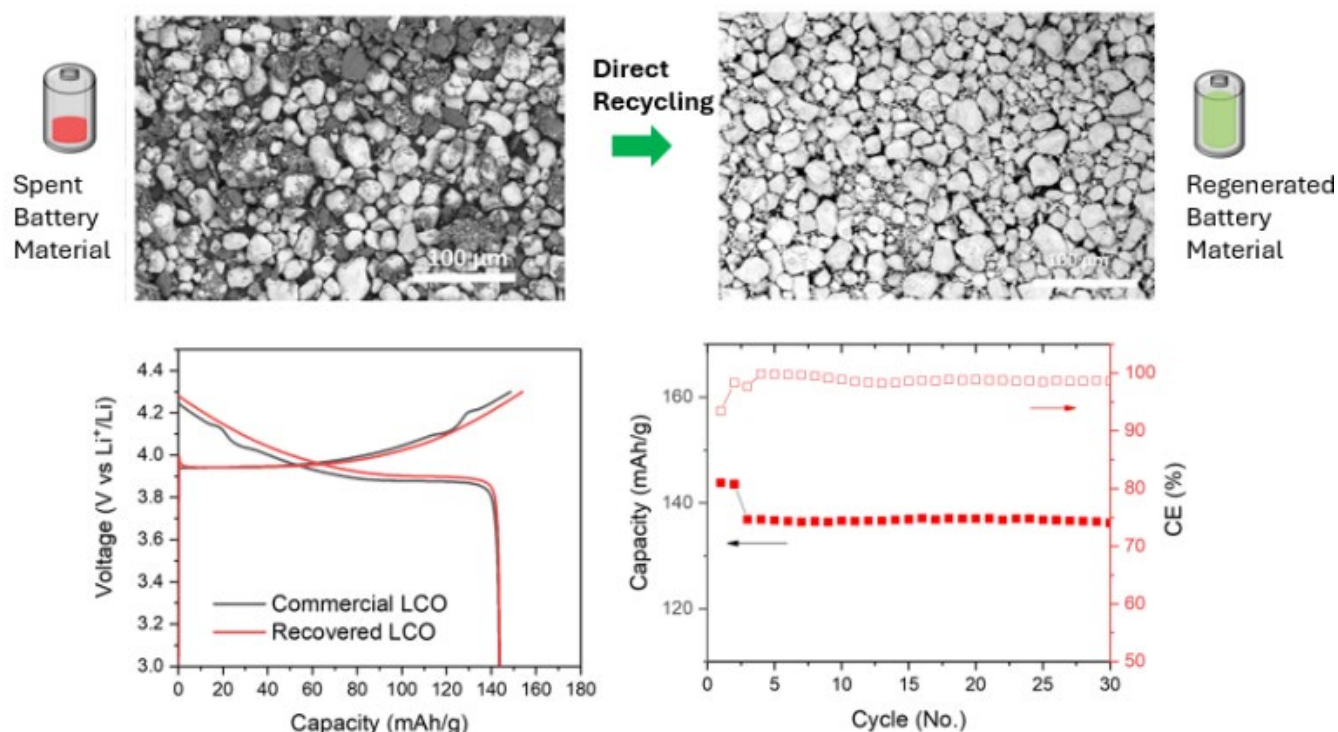
The project successfully achieved all key performance metrics outlined for the set tasks. These include:

- The development of a methodology and process for the deactivation, dismantling, and separation of components from spent LIBs, ensuring high safety standards and low energy costs.
- The establishment of an operational procedure detailing the methodology and protocols for extracting over 90 percent of the cathode and anode active materials from spent batteries, compared to the expected theoretical value.
- The successful application of a froth floatation aqueous technique for separation, which is both environmentally friendly and industrially mature. The purity of the value-concentrated cathode black mass obtained through this protocol exceeded 97 percent, making it suitable for the direct regeneration process.

The newly developed PRIME regeneration process significantly enhanced the efficiency of direct recycling by incorporating impurity removal preprocessing steps within the regeneration protocol. The final cathode products with various chemistries such as LCO, NCM111, and NCM622 recovered through the PRIME process demonstrated high quality, matching the characteristic features and performance of commercial samples.

Cell fabrication and electrochemical testing were conducted at UCSD's advanced facilities, with initial cell assembly into coin cells and electrochemical evaluations performed at the SPEC Center. The recovered LCO cathode, processed via froth flotation, was tested in coin cells using lithium chips as counter electrodes. As shown in Figure 16, the recovered LCO CAMs demonstrated a first-cycle discharge capacity of 143 mAh/g with a CE of 93.46 percent. For comparison, virgin materials displayed a similar discharge capacity with a slightly higher CE, indicating close to 100 percent regeneration. These findings confirm the high quality of the recovered CAMs, with impurity levels effectively managed throughout the process. Figure 17 further highlights the cycling stability of the recovered CAMs in half cells, showcasing consistent performance over multiple cycles. Notably, this represents UCSD's first successful demonstration of a complete end-to-end process for recovering and regenerating CAMs.

Figure 16: Morphology and Electrochemical Performance of the Recycled LCO Sample From LIBs



Source: UCSD

Similar results were achieved for other CAMs with varying morphologies and more complex chemistries, such as NCM111 and NCM622. Characterizations highlighting crystal structure and stoichiometric issues in CBM are detailed in Table 5 and Figure 17. Figure 20a, Figure 20b, and Figure 20c present SEM images of the NCM111 CBM, the CBM after hydrothermal treatment and washing, and the recovered CAM (rCAM), respectively. The SEM image of the CBM clearly indicates that impurities, such as binder and carbon, covered most of the cathode surface.

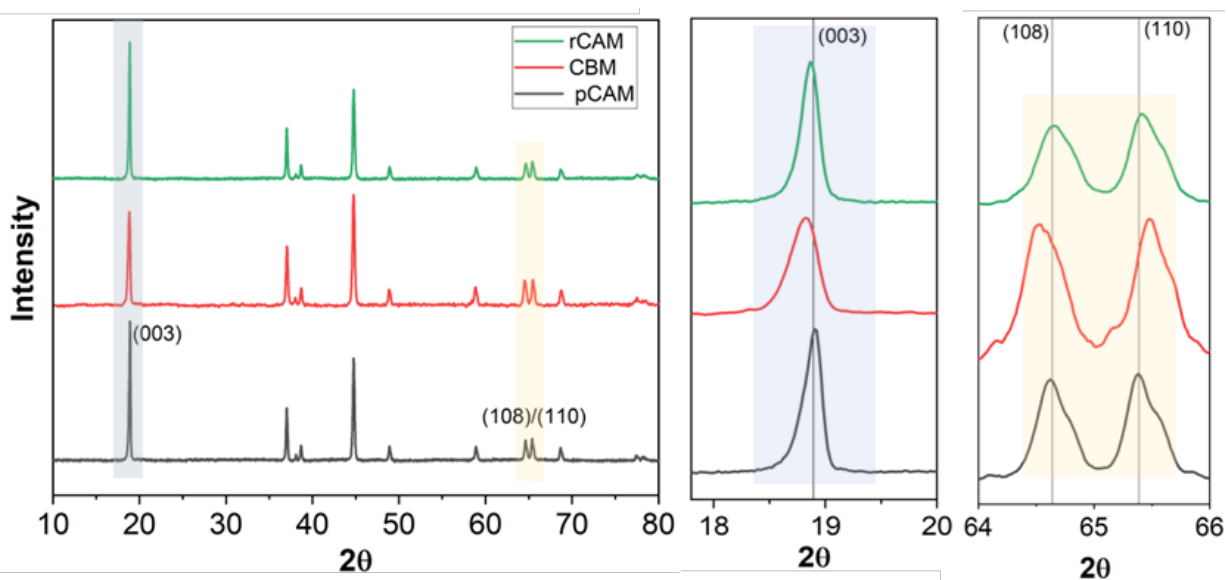
Table 5: NCM111 and NCM622 ICP-MS Data Showing Relithiation in Recycled Cathode

	Li/TM ratio	Ni/TM ratio	Co/TM ratio	Mn/TM ratio
Pristine NCM111	1.08	0.34	0.34	0.32
CBM NCM111	0.89	0.36	0.30	0.34
Recycled cathode (NCM111)	1.02	0.36	0.30	0.34
Pristine NCM622	1.05	0.60	0.20	0.20
CBM NCM622	0.91	0.61	0.20	0.19
Recycled cathode (NCM622)	1.07	0.61	0.20	0.19

Key: Li=lithium, Ni=nickel, Co=cobalt, Mn=manganese, TM=transition metal.

Source: UCSD

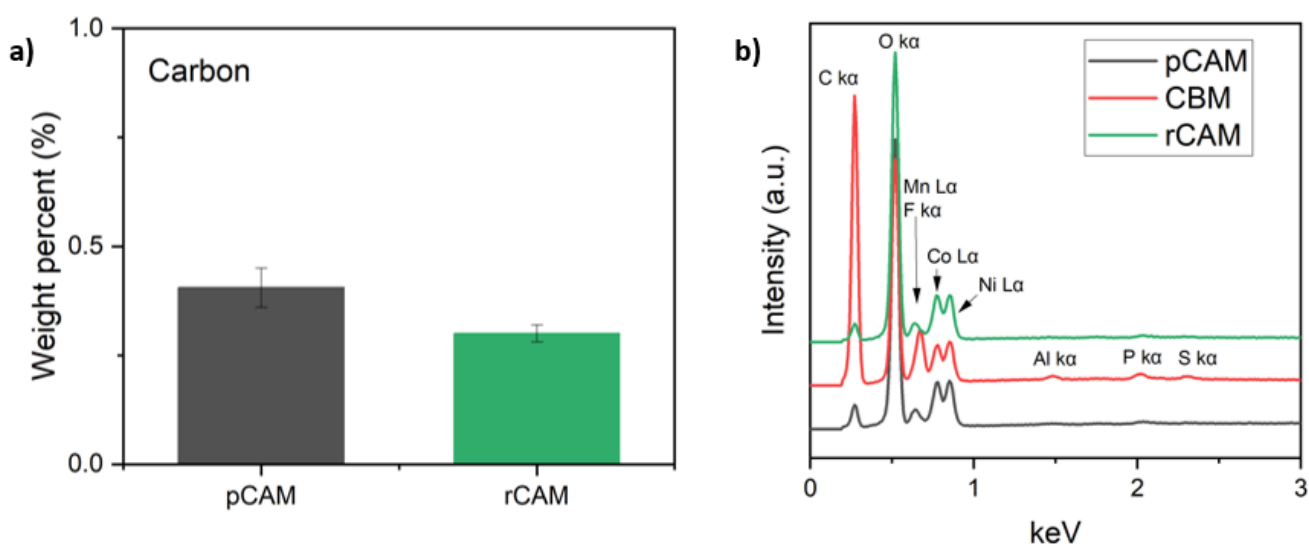
Figure 17: Crystal Structure Evolution of Recycled NCM111 CAM (rCAM) from CBM Compared to Pristine NCM111 Material (pCAM)



Source: UCSD

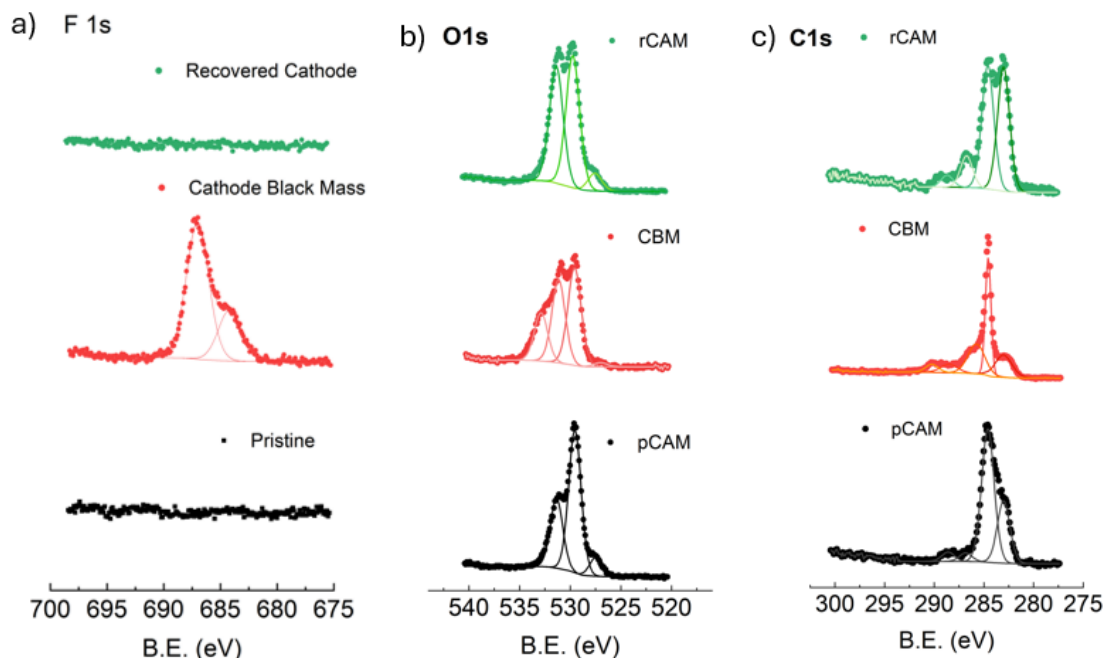
These impurities were effectively removed through the recycling steps, as evidenced by the rCAM's clean surface, demonstrating the success of the purification process. Additional analyses, including impurity tracking and end-product quality assessments, are also provided in Figure 17, Figure 18, and Figure 19. The recovered materials achieved over 99 percent purity, comparable to pristine commercial counterparts.

Figure 18: Impurity Analysis of (a) Carbon by Combustion Analysis and (b) Aluminum by EDS, for Quality Comparison Between Recycled NCM111 CAM (rCAM) and Pristine NCM111 Material (pCAM)



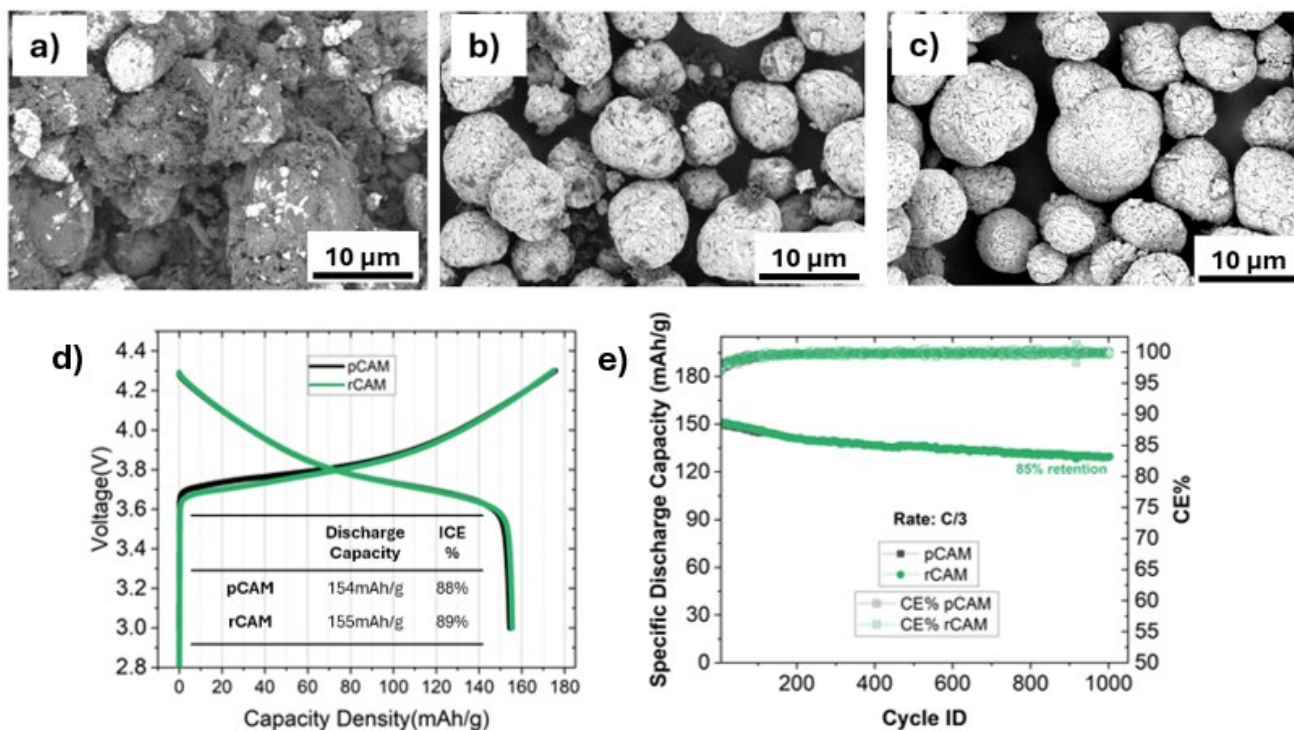
Source: UCSD

Figure 19: XPS* Data of (a) Fluorine, (b) Oxygen and (c) Carbon to Indicate Removal of the Binder and Cathode Electrolyte Interphase From the Recycled Cathode (rCAM)



*XPS=X-ray photoelectron spectroscopy.
Source: UCSD

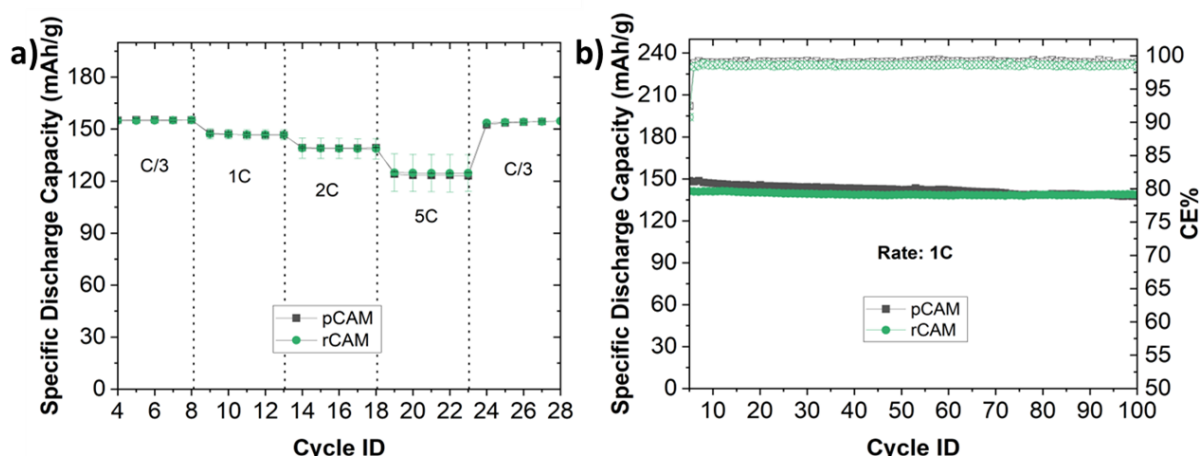
Figure 20: Morphology and Electrochemical Performance of the Recycled NCM111 Sample From Spent LIBs



Source: UCSD

After confirming impurity removal and rCAM quality, electrochemical performance was evaluated using half-cell and full-cell configurations. The voltage profile for the first cycle at a C/10 rate (Figure 20d) shows a comparable discharge capacity of 155 mAh g⁻¹ for the rCAM and 154 mAh g⁻¹ for the pCAM. The initial Coulombic efficiency (ICE) of the rCAM was 89 percent, closely matching the 88 percent observed for the pCAM. Half-cell cycling at a C/3 rate demonstrated 98 percent discharge capacity retention after 100 cycles for the rCAM, similar to the pCAM. Longer cycling over 1,000 cycles retained 85 percent discharge capacity (Figure 20e), attesting to the high quality of the rCAM. Rate performance evaluation further confirmed that the rCAM matched well with the pCAM, validating the effectiveness of the direct recycling process (Figure 21). Additionally, full-cell configurations using graphite as the anode exhibited similar trends, achieving capacity retention greater than 94 percent at a 1C rate after 100 cycles for both the pCAM and the rCAM (Figure 21b). These results demonstrate that the rCAM exhibits electrochemical performance on a par with the pCAM in all aspects.

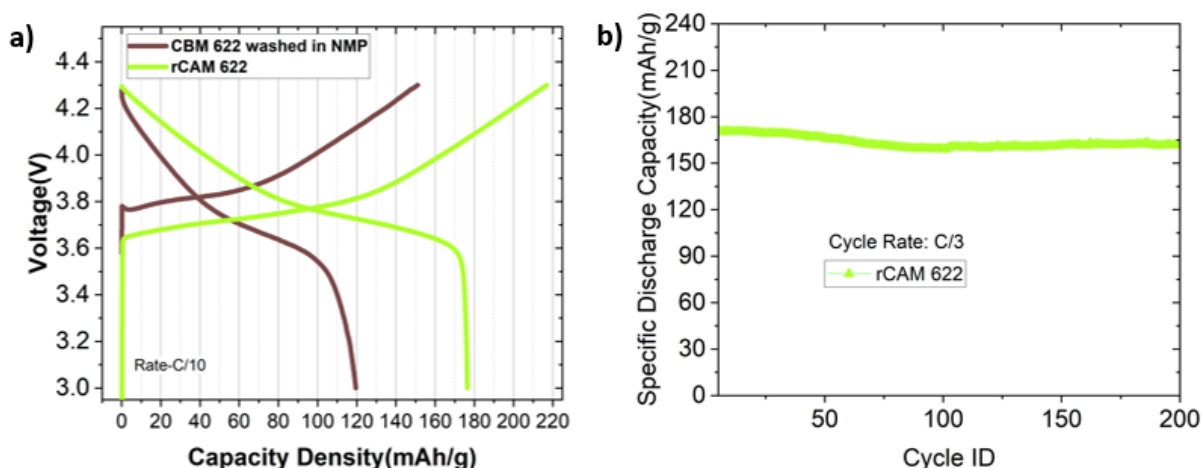
Figure 21: Electrochemical Performance of the Recycled NCM111 CAM (rCAM) and Pristine NCM111 Material (pCAM): a) Rate Performance of the Regenerated Cathode Compared With Pristine, and b) Cycling Stability in Full Cell



Source: UCSD

Further, CBM obtained from NCM622 cells (CBM622) was treated using the same process as for NCM111 CBM, except for the annealing step, which was performed under pure oxygen flow rather than air to preserve the sensitive Ni-rich chemistry. The first-cycle voltage profiles for the recovered NCM622 cathode (rCAM622) and a control CBM622 sample washed in N-Methyl-2-pyrrolidone (NMP) are compared in Figure 22a. The ICE of the CBM622 half-cell was 79 percent, likely due to the presence of impurities, with a discharge capacity of only 119 mAh g⁻¹. In contrast, the rCAM622 half-cell exhibited an ICE of 82 percent with a discharge capacity of 176 mAh g⁻¹, matching commercial-level NCM622 performance. Long-term half-cell cycling data (Figure 22b) show 94-percent capacity retention after 200 cycles, indicating the high quality of the regenerated cathode material.

Figure 22: Electrochemical Performance of the Recovered NCM622 Cathode (rCAM622) and Control CBM622: (a) First-cycle Voltage Profile and (b) Cycling Stability

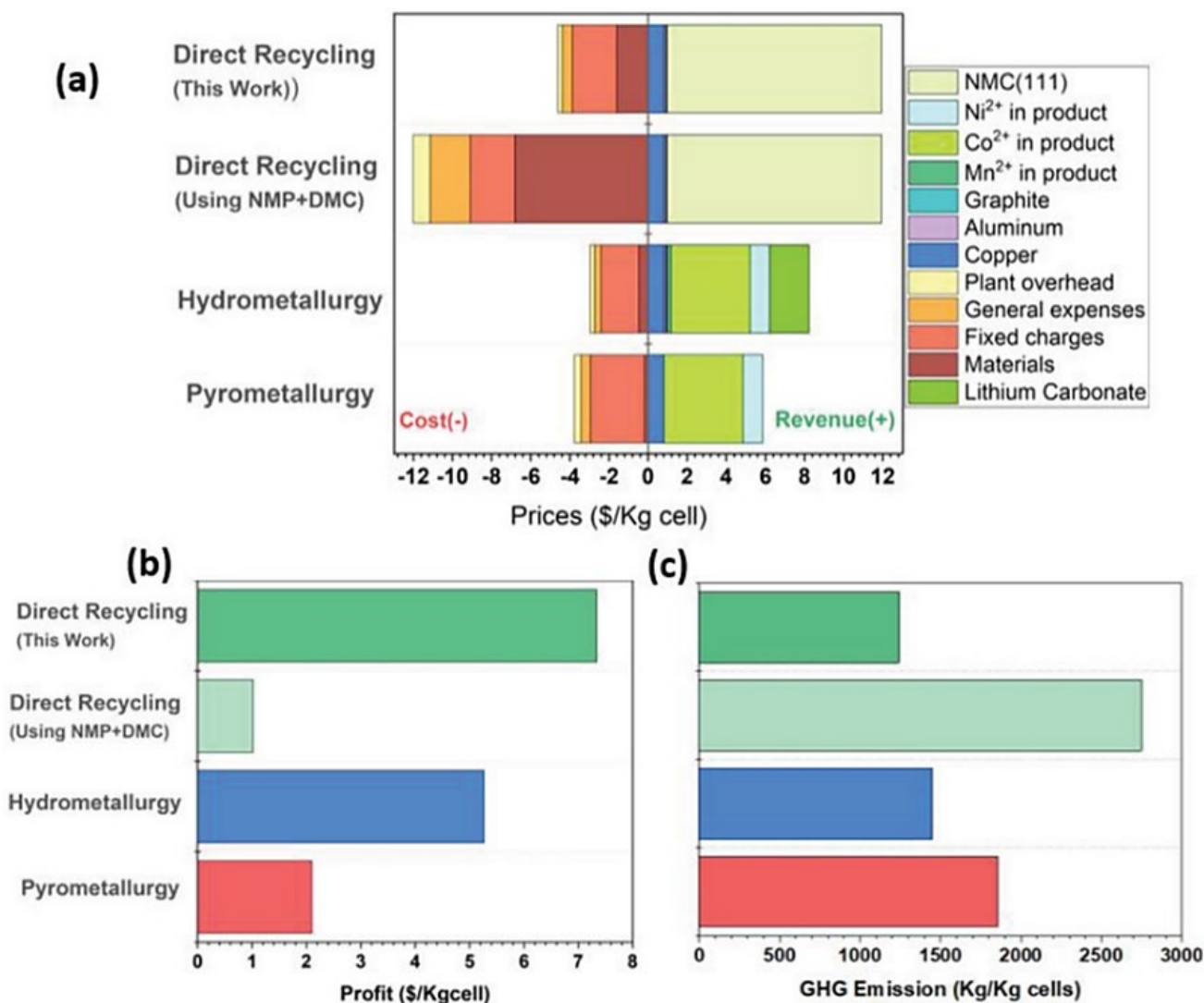


Source: UCSD

These results demonstrate that the PRIME process is robust and resilient, effectively handling up to 10 percent reductive carbon impurities in the CBM feedstock without compromising performance.

Most existing direct recycling methods rely on NMP or similarly toxic organic solvents to remove PVDF, while other approaches use dimethyl carbonate (DMC) to wash electrolyte salts. The widespread use of these solvents necessitates their inclusion in economic and environmental analyses for scaling up, a consideration often overlooked in many studies. To address this gap, a comparative analysis of traditional pyrometallurgical recycling, current hydrometallurgical techniques, the conventional laboratory-scale direct recycling process (employing NMP and DMC), and the novel PRIME direct recycling benchmark was conducted. As shown in Figure 23, the process eliminates the use of expensive organic solvents, reducing material costs by approximately \$5/kg of battery cells (Figure 23a). Additionally, avoiding organic solvents significantly lowers general expenses and plant overhead costs, including labor, supervision, administration, and maintenance, resulting in further savings of approximately \$2.16/kg of battery cells. The analysis highlights that direct recycling methods relying on NMP and DMC are less economically viable than even the current pyrometallurgical and hydrometallurgical approaches (Figure 23b). Moreover, the industrial scalability of such solvent-based methods is restricted due to global regulations limiting the use of toxic solvents. The environmental implications are equally compelling. Figure 23c demonstrates that the greenhouse gas emissions associated with NMP- and DMC-based direct recycling exceed those of the traditionally pollutive pyrometallurgical processes.

Figure 23: Comparison of Various Recycling Method Calculated Using Everbatt Model



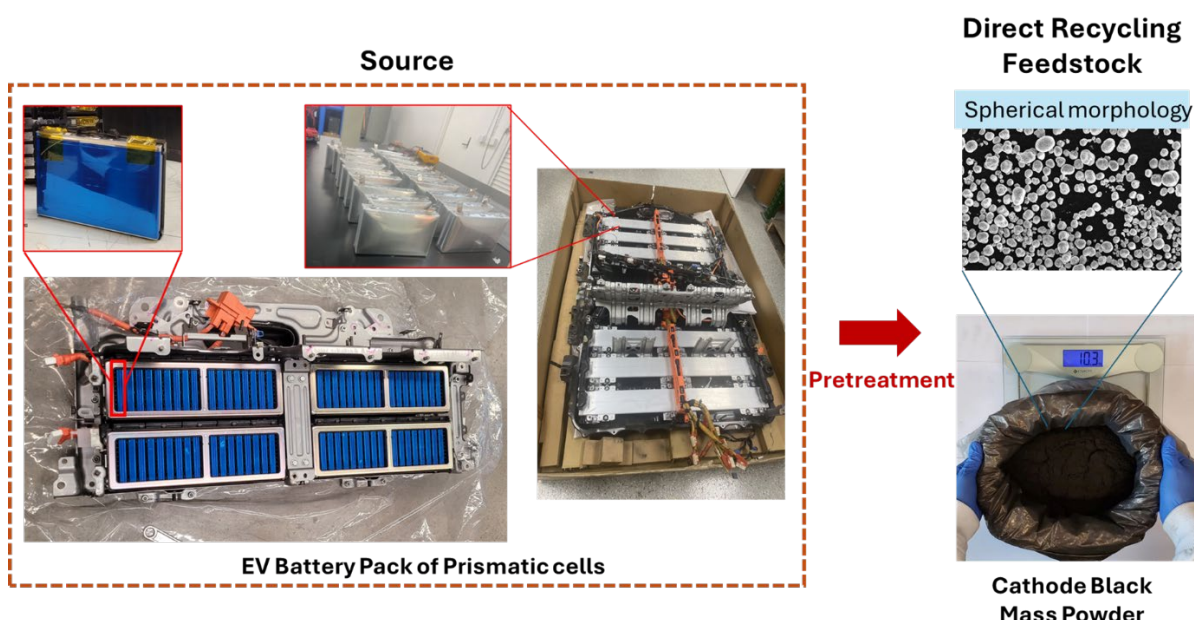
Source: UCSD

More specifically, the PRIME process reduces greenhouse gas emissions for recycling a kilogram of spent cells, from 2,746 kg for conventional direct recycling to 1,243 kg. This is even lower than conventional hydrometallurgy and pyrometallurgical recycling methods, which emit 1,449 kg and 1,852 kg, respectively, of greenhouse gases per kilogram of spent cells. By removing these economic and environmental barriers, the PRIME direct recycling process not only aligns with environmental sustainability at scale but also enhances profitability.

A pilot-scale demonstration was conducted to regenerate 5 kg of cathode active material from spent EV battery packs, validating the scalability and industrial relevance of the developed technology. Figure 24 presents a schematic of the CBM recovery process from the received battery packs and modules using the pretreatment protocol established in this project. Figure 25 provides a visual representation of the scaled-up CBM processing through the PRIME

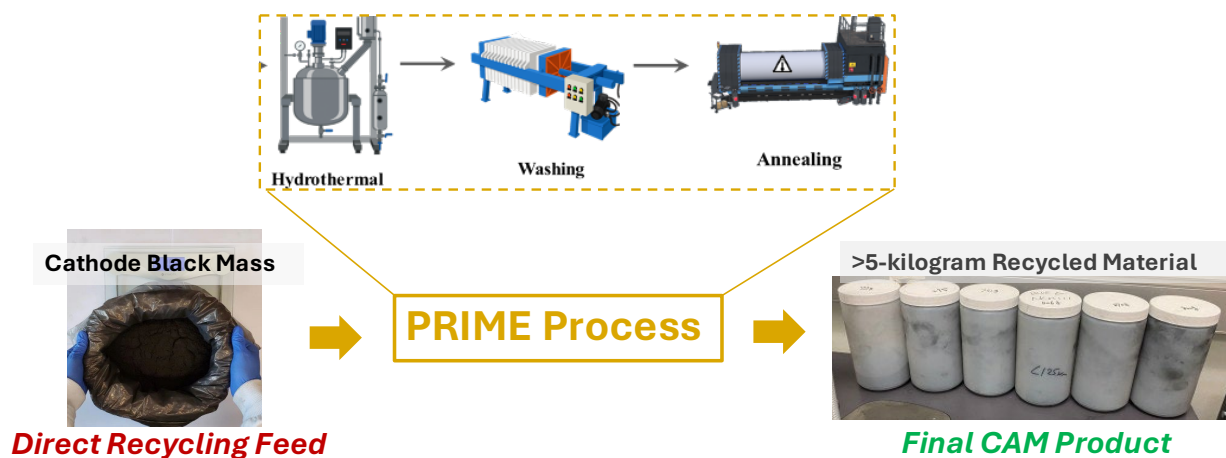
regeneration process. The regenerated materials obtained at pilot scale exhibited performance and quality comparable to those produced in earlier laboratory-scale trials.

Figure 24: Scaled-up Feedstock Recovery From Spent EV Battery Packs Using the Pretreatment Protocol Developed in This Project



Source: UCSD

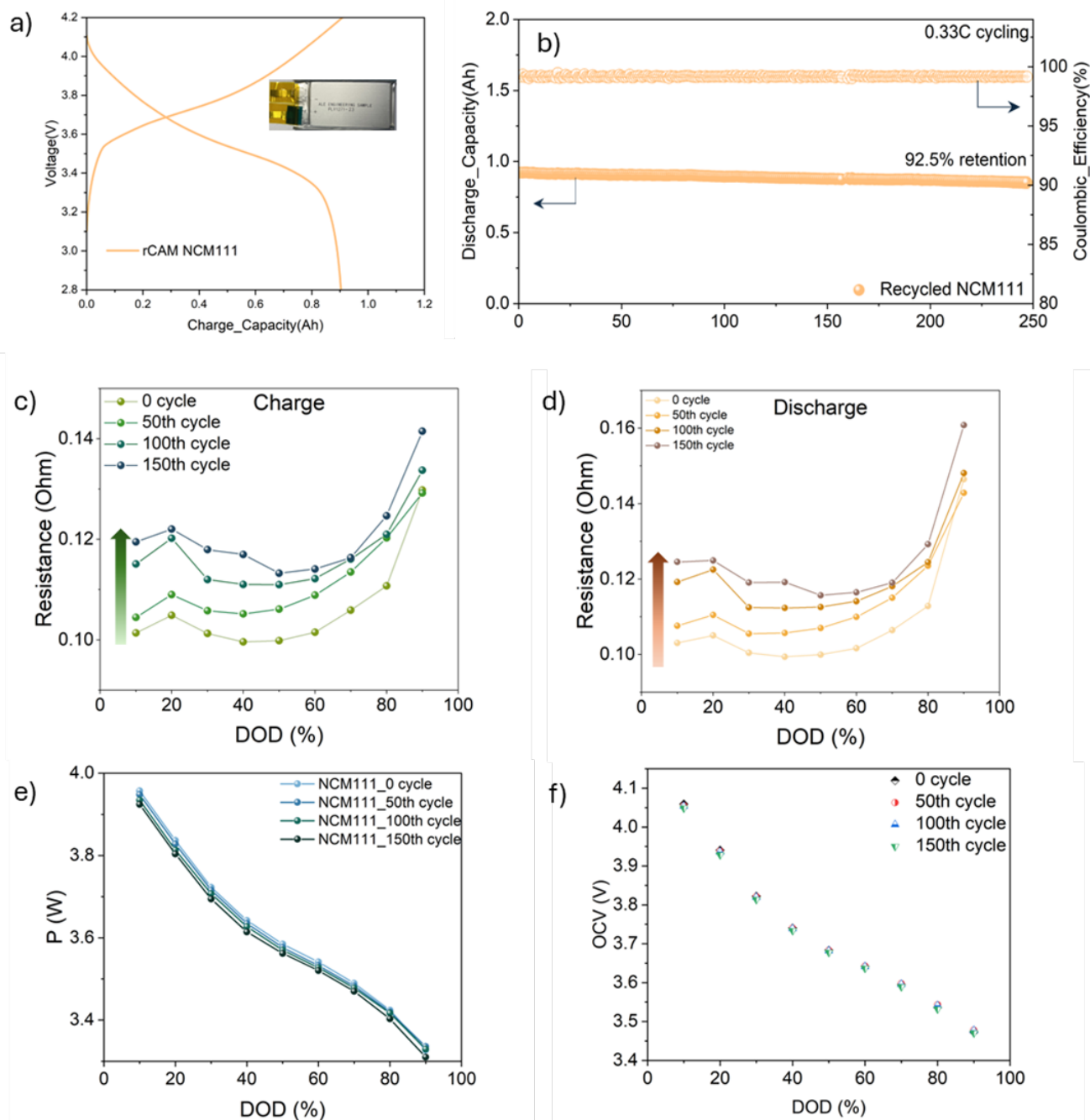
Figure 25: Demonstration of Direct Recycling Scale-up via the PRIME Process Developed in This Project



Source: UCSD

To further assess their real-world performance, 1-Ah multilayer pouch cells were fabricated using the regenerated NCM111 material at ALE's state-of-the-art facility. These cells, shown in the inset of Figure 26, were evaluated at UCSD across various performance metrics, including cycle life, impedance rise, power fade, and CE.

Figure 26: Pouch Cell Testing of rCAM NCM111 for Electrochemical, Power, and Impedance Performance Verification

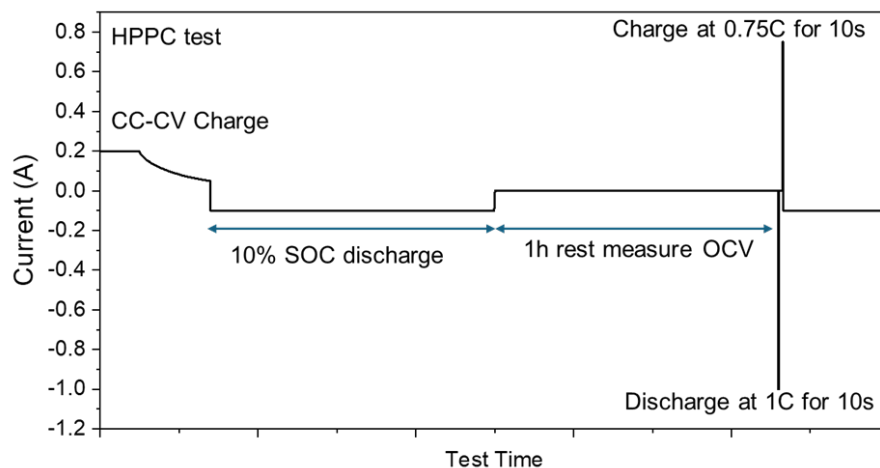


Source: UCSD

The pouch cells were cycled at 0.33C within a voltage window of 3.0–4.2 volts, delivering an average capacity of 0.92 Ah with an ICE of 99.2 percent and a capacity retention of 92.5 percent after 250 cycles (Figure 26b), indicating excellent bulk material stability and high electrochemical performance of the regenerated cathode. To further assess dynamic behavior, hybrid pulse power characterization (HPPC) testing was performed to evaluate power

capability and track internal resistance evolution under load (the testing protocol shown in Figure 27).

Figure 27: HPPC Testing Protocol Used to Test Impedance and Power Performance of Pouch Cells



Source: UCSD

As shown in Figure 26c, internal resistance during charging increased with depth of discharge (DOD), consistent with the expected behavior of NCM materials. However, when comparing the 0th and the 150th cycles, a noticeable increase in resistance was observed, particularly at lower DOD percentages. This rise is attributed to interfacial buildup at the electrode-electrolyte interface, leading to localized impedance growth. Despite this, the strong capacity retention suggests that these effects are surface-limited and do not significantly hinder lithium accessibility or active material utilization. A similar trend was observed for discharge resistance as a function of DOD percentage (Figure 26d).

Power output and open-circuit voltage decreased with an increase of DOD percentage, as expected due to rising resistance. However, comparisons over 150 cycles (Figure 26e and Figure 26f) showed consistent trends, indicating that the increased resistance at low DOD has minimal impact on overall power performance. These results confirm that the regenerated cathode material maintains expected impedance and power characteristics under practical cycling conditions. Overall, the data demonstrates that the recycled NCM111 performs comparably to pristine material in terms of capacity, efficiency, and dynamic power response.

These larger-scale evaluations verified the consistency and efficacy of the regeneration methodologies. The data collected provided a comprehensive performance profile, highlighting advancements in cathode chemistries achieved through this program and underscoring the successful collaboration among the project partners. The research conducted under this project culminated in two peer-reviewed publications: *Scalable Direct Recycling of Cathode Black Mass from Spent Lithium-Ion Batteries* and *Direct Recycling Industrialization of Li-Ion Batteries: The Pre-Processing Barricade*. Additionally, the PRIME process and related findings were presented at the Electrochemical Society's 2023 conference, where they attracted significant interest from the scientific and industrial communities.

CHAPTER 4:

Technology and Knowledge Transfer

The project team's objective with technology and knowledge transfer was to build upon the knowledge gained from the direct recycling process and implement activities that would accelerate the commercial adoption of the technology. A key step in this transition was the successful transfer of the PRIME process, a low-temperature, solution-based direct recycling method developed at UCSD, to ExPost Technology, a UCSD spin-off company based in San Diego.

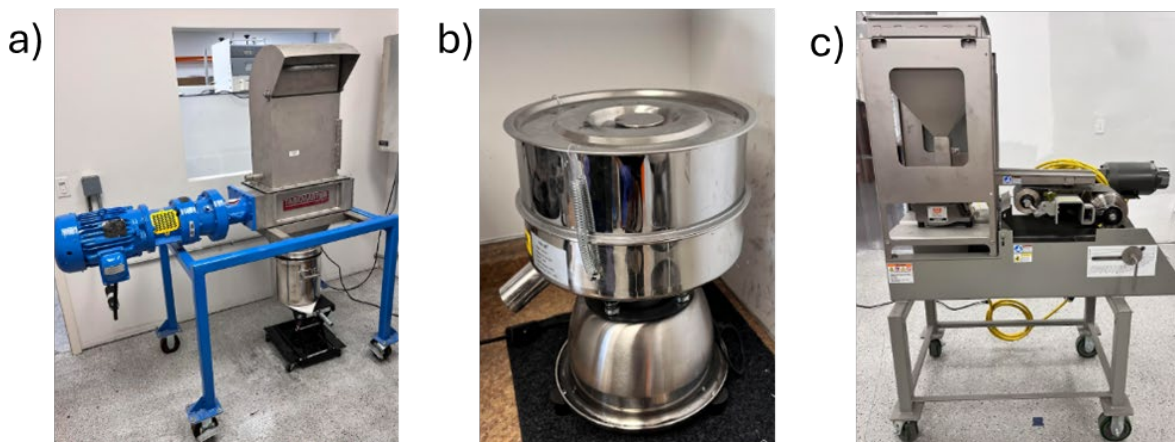
ExPost Technology is focused on industrializing the direct recycling of LIBs by scaling up the PRIME process and validating its commercial viability. The PRIME process, patented by UCSD during the course of this project, was licensed to ExPost for commercialization. The UCSD team provided in-depth technical training and documentation to ensure that ExPost could reproduce and further advance the process at a larger scale. This collaborative effort marked a significant step toward market readiness.

ExPost has made substantial progress in adapting and scaling the PRIME process for industrial application. One of the major accomplishments was the installation and operation of multiple pre-pilot systems at its facility, which now supports streamlined processing of larger volumes of spent battery materials. Each stage of UCSD's laboratory-scale process was optimized at the multi-kilogram scale, ranging from feedstock preparation to final product recovery, with a focus on maintaining high material recovery efficiency and consistent performance.

A series of specialized preprocessing equipment installations enabled ExPost to transition from laboratory-scale to pre-pilot-scale operations. These include a high-capacity shredder capable of processing over 10 kg of battery feedstock per hour, as shown in Figure 28a. This unit effectively produces uniform shredded material in the 1–5 mm range, with a narrow particle size distribution suitable for downstream separation.

Following shredding, the material is sieved to isolate the valuable black mass from casing materials and current collectors, including aluminum, copper, and plastic. ExPost employs shaking sieving equipment that operates at a throughput of approximately 1 kg/hr, as illustrated in Figure 28b. To further purify the black mass, magnetic separation is applied to remove steel-based magnetic impurities. This operation, depicted in Figure 26c, achieves processing scales of 5–10 kg/hr, depending on the input characteristics.

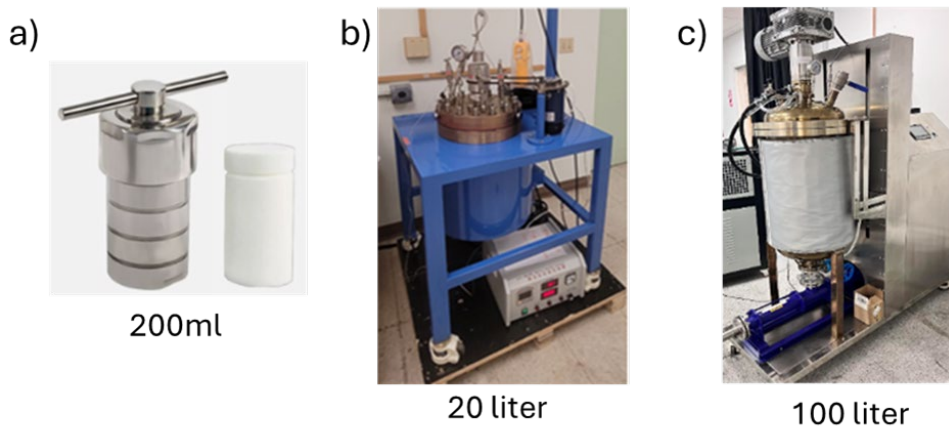
Figure 28: Scaled Preprocessing Equipment



Source: ExPost Technology

In parallel with these mechanical preprocessing operations, ExPost also significantly scaled the wet-chemical processing portion of the PRIME process. The PRIME process operates at temperatures up to 302°F (150°C). Initial relithiation experiments were conducted at the UCSD laboratory using small-scale hydrothermal reactors ranging from 200 mL to 1 liter in volume (Figure 29a). As the process matured, it was scaled up to a 20L reactor at UCSD (Figure 29b). More recently, a 100L reactor capable of processing 10–20 kg per batch was designed and installed by ExPost (Figure 29c), marking a significant milestone toward industrial-scale implementation. This upgrade is critical for improving throughput and enabling the process to meet commercial production demands efficiently.

Figure 29: Scaled-up PRIME Reactor Size

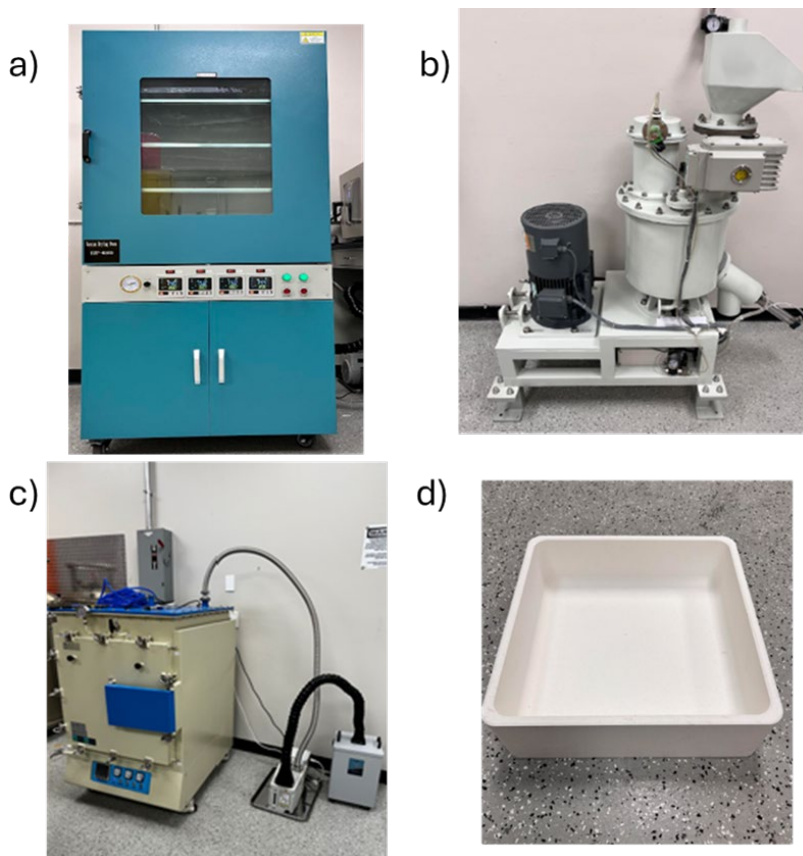


Source: ExPost Technology

Additional advancements at ExPost include the integration of critical post-PRIME processing steps necessary to restore the performance of regenerated cathode materials. These steps include vacuum drying, high-speed mixing, and sintering in an atmosphere-controlled furnace. After the PRIME process, the material is thoroughly dried and mixed with lithium hydroxide, using a vacuum drying oven with a 430-liter capacity (Figure 30a) and a high-speed mixer with

a throughput of 5 kg/hr (Figure 30b). This step is essential for achieving uniform lithium distribution within the cathode material, effectively compensating for lithium loss that may have occurred during earlier processing stages.

Figure 30: Scaled-up Post-PRIME Processing



Source: ExPost Technology

To validate large-scale operation, ExPost also established sintering capabilities using a high-capacity, atmosphere-controlled box furnace capable of processing more than 10 kg per batch (Figure 30c). This sintering step improves the structural integrity and electrochemical performance of the cathode active material. To support batch scalability, industrial-grade saggars, each capable of holding at least 5 kg of material, were procured, and the furnace is designed to accommodate multiple saggars in a single run (Figure 30d).

The successful technology transfer, equipment commissioning, and process validation conducted during this project significantly advanced the commercialization potential of the PRIME process. With ExPost Technology now operating as an independent entity capable of pre-pilot production, this work effectively laid the foundation for full industrial-scale deployment of direct LIB recycling in California.

CHAPTER 5:

Conclusion

The project achieved significant milestones in advancing sustainable and efficient direct recycling technologies for spent LIBs. It successfully developed cost-effective sorting and de-energization processes, ensuring the safe and efficient separation of LIB components. By leveraging innovative interfacial chemistry tuning, the team optimized a scalable and environmentally friendly aqueous separation method to recover high-purity cathode and anode materials. Additionally, the integration of purification and regeneration processes was accomplished using multipurpose regeneration conditions, effectively bypassing the need for toxic organic solvents. A robust process design strategy was also established, laying the foundation for the commercialization of direct recycling technologies for EV and energy storage system (ESS) batteries.

Benefits to Ratepayers

As highlighted in Chapter 3, the PRIME process has been shown to greatly decrease energy consumption, the utilization and need for expensive and toxic chemicals, and overall greenhouse gas emissions associated with the processing and recycling of lithium batteries and cathode/anode materials. Consequently, the scale-up and commercialization of PRIME will result in significant benefits to California ratepayers.

Lithium-ion batteries are critical to achieving decarbonization in both the transportation and power sectors. The state projects that it will need 52,000 megawatts (MW) (208,000 megawatt-hours [MWh] over 4 hours) of battery storage by 2045 (California Energy Commission, April 2025). Reducing the energy, chemical, greenhouse gas, and costs of battery recycling using technologies such as PRIME at scale will result in significant benefits to ratepayers. This can take the form of reduced energy storage procurement costs, supply chain reinforcement, and lower lifecycle impacts to sustain the transition to renewable energy and EVs, which can help drive both downward pressure on rates and greater environmental sustainability.

Table 6 summarizes benefit projections, comparing direct recycling with pyro and hydro processes, assuming an estimated 45,000 battery packs that will be retired annually by 2027. Assuming 75 percent of these packs, with an average capacity of 50 kilowatt-hours (kWh), require recycling (with the other 25 percent available for reuse) and assuming that the stationary battery storage market is equivalent to 20 percent of the transportation sector, the total battery volume in California available for recycling will be 10 million kg (5 kg per kWh) in 2027. Estimates of total recycled volumes in 2030, 2035, and 2040 were projected based on an annual 15-percent growth in volume. Using the quantified metrics in Chapter 4 for energy consumption and recycling costs, the following figures are calculated for annual energy, economic, and GHG savings:

**Table 6: Benefit Projections Comparing Direct Recycling
with Pyro and Hydro Processes**

Energy Consumption (Annual GWh)	2030	2035	2040
Pyro	870 GWh	1,980 GWh	4,100 GWh
Hydro	1,450 GWh	3,330 GWh	6,850 GWh
Direct	165 GWh	380 GWh	760 GWh
Annual Energy Savings From Direct	705-1,285 GWh	1,600-2,950 GWh	3,340-6,090 GWh
Recycling Costs (Annual \$)			
Pyro	\$44.9M	\$102M	\$210M
Hydro	\$31.7M	\$72.0M	\$148M
Direct	\$27.7M	\$63.0M	\$130M
Annual Cost Savings From Direct	\$4M - \$17M	\$9M-\$39M	\$19M-\$81M
Annual GHG Reductions* From Direct Compared to Pyro and Hydro	(80,000 – 140,000) MT	(100,000 – 190,000) MT	(67,000 – 120,000) MT

***Assumes 200 kg/MWh (EIA) emissions in 2019, decreasing to 20 kg/MWh by 2040.**

Key: GWh=gigawatt-hours, M=million, MT= metric tons
Source: UCSD

Potential of Demonstrated Technology

The project successfully achieved its goal of developing a comprehensive technology transfer plan, leading to the commercialization of direct recycling technology. UCSD obtained a patent for its PRIME technology and established a pilot direct recycling facility with ExPost Technology, to demonstrate scalability. Additionally, UCSD secured \$10 million in DOE Bipartisan Infrastructure Law funding to scale up the technology to the 100-kg level by the project's conclusion. Leveraging its extensive network of partners from previous CEC and DOE programs, the project team devised a robust marketing strategy for early market access. This included obtaining relevant certifications to meet industry standards, paving the way for large-scale production and widespread adoption.

Need for Future Support

UCSD has strategic partnerships with battery recyclers, automakers, DOE labs, and investor-owned utilities that will firm the value proposition for the direct recycling of retired EV batteries and used utility-scale batteries. The team will seek follow-up funding from federal, state, and private sources to advance its direct recycling technology to TRL 7.

Future Research Recommendations

For future research work, the project team recommends focusing on developing physical and dry separation techniques, since they are usually nondisruptive to CAM chemistry and have lower waste generation. Eventually, processes with the lowest cost, lowest carbon footprint, and highest quality feedstock will be adopted in industry; therefore, these factors should be the core of the process design. California's clean energy policies have created a large and growing market for EVs and ESS. Low-impact and economically attractive recycling technologies are critical to establishing in-state battery recycling capacity that can process this growing electronic waste stream while reducing the high costs of transporting EoL batteries to out-of-state facilities.

GLOSSARY AND LIST OF ACRONYMS

Term	Definition
AAM	anode active material
ABM	anode black mass
Ah	ampere-hour
ALE	American Lithium Energy Corporation
CAM	cathode active material
CBM	cathode black mass
CE	Coulombic efficiency
CEC	California Energy Commission
cm ⁻¹	inverse centimeters
CoSO ₄	cobalt sulfate
Cu	copper
DCIR	Direct Current Internal Resistance
DMC	dimethyl carbonate
DOD	depth of discharge
DOE	U.S. Department of Energy
EDS	energy dispersive X-ray spectroscopy
EOL	end-of-life
ESS	energy storage system
EV	electric vehicle
ExPost	Expost Technology
FTIR	Fourier transform infrared spectroscopy
GHG	greenhouse gas
GW	gigawatts
GWh	gigawatt-hours
HPPC	hybrid pulse power characterization
hydro	hydrometallurgical
H ₂ O ₂	hydrogen peroxide
H ₂ SO ₄	sulfuric acid
ICE	initial Coulombic efficiency
ICP-MS	inductively coupled plasma mass spectroscopy
Kg	kilograms

Term	Definition
kWh	kilowatt-hours
LCO	lithium cobalt oxide
Li	lithium
LIBS	lithium-ion batteries
LIOH	lithium hydroxide
LFP	lithium iron phosphate
LMO	lithium manganese oxide
Li ₂ CO ₃	lithium carbonate
mAh/g	milliampere-hours per gram
mL	milliliter
MW	megawatt
MWh	megawatt-hours
NCA	nickel cobalt aluminum
NCM	lithium nickel cobalt manganese oxide
NMC	nickel manganese cobalt
NMP	N-Methyl-2-pyrrolidone
NiSO ₄	nickel sulfate
NSF	National Science Foundation
PRIME	Purification and Regeneration Integrated Materials Engineering
PCAM	pristine cathode active material
PVDF	polyvinylidene fluoride
Pyro	pyrometallurgical
rAAM	Recycled Anode Active Material
rCAM	recycled cathode active material
rCAM-G2	recycled cathode active material-generation 2 (product from second re-use of lithium supplement solution)
SEM	scanning electron microscopy
SOC	state of charge
SOH	state of health
SPEC	Sustainable Power and Energy Center
S/L	solid-to-liquid (ratio)
TRL	Technology Readiness Level
UCSD	University of California, San Diego

Term	Definition
XPS	X-ray photoelectron spectroscopy
XRD	X-ray diffraction
μm	micrometer

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Project Deliverables

Project deliverables, including interim project reports, are available upon request by submitting an email to pubs@energy.ca.gov. This includes the following:

- Final Battery Rapid Integration Report
- Final Separation Operations Procedure Report
- Final Electrode Direct Regeneration Procedure Report
- Final Process Scale-Up Report
- Final Cell Fabrication Report
- Final Electrochemical Measurement Report
- Final Healing Chemistry Report
- Final Report on Demonstration of Recycled Battery Electrode in DAC Vehicle Fleet
- Final Life Cycle Energy Use and GHG Emissions Report
- Final Life Cycle Costs and Revenues Report
- Final Technology Transfer Plan
- Final Technology Transfer Summary Report



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APPENDIX A: Relevant Protocols, Calculations, Materials Structure, Composition, and Electrochemical Property Data

October 2025 | CEC-500-2025-044

APPENDIX A:

Relevant Protocols, Calculations, Materials Structure, Composition, and Electrochemical Property Data

Protocol for State of Health (SOH) Assessment of LIBs

The project team employed a combination of two aging variables to assess SOH. By analyzing these parameters, we can evaluate the battery's overall health and determine its suitability for further use or recycling. Specifically, the pulse discharge test is used to measure two key parameters:

- **Internal Resistance (DCIR):** DCIR, or Direct Current Internal Resistance, measures the resistance within a battery that opposes the flow of electrical current. A higher DCIR indicates increased internal resistance, leading to reduced performance.

Steps to Calculate DCIR (Direct Current Internal Resistance) from a Pulse Discharge Test:

1. Measure the Current and Voltage Drop:
 - During pulse, measure the current (typically constant during the pulse).
 - Measure the voltage drop across the battery during the pulse discharge $\Delta V = V_{\text{start}} - V_{\text{end}}$
 - V_{start} is the voltage at the beginning of the pulse, and V_{end} is the voltage at the end of the pulse.
 2. Calculate the DCIR:
 - The DCIR is calculated using Ohm's Law: $\text{DCIR} = \Delta V / I$
- **Remaining Capacity:** The total capacity delivered during the discharge test provides insight into the battery's remaining usable capacity.

Steps to Calculate Remaining Capacity from the Pulse Discharge Test:

1. Measure the Total Energy Delivered:
 - During the pulse test, measure the total energy or charge delivered (in Ampere-hours or Ah). This can be done by integrating the current over the discharge time: $\text{Energy Delivered (Ah)} = \int I(t)dt$
 - This integral gives the total charge (in Ah) discharged by the battery.
2. Estimate Remaining Capacity:
 - The remaining capacity is calculated as: $\text{Remaining Capacity (Ah)} = \text{Energy Delivered (Ah)} / \Delta\text{SOC}$

- Where, ΔSOC is the difference in the State of Charge before and after the pulse discharge.

By analyzing these parameters, the battery's overall health can be evaluated to determine its suitability for further use or recycling.

• Overall State of Health (SOH) Calculation and Evaluation

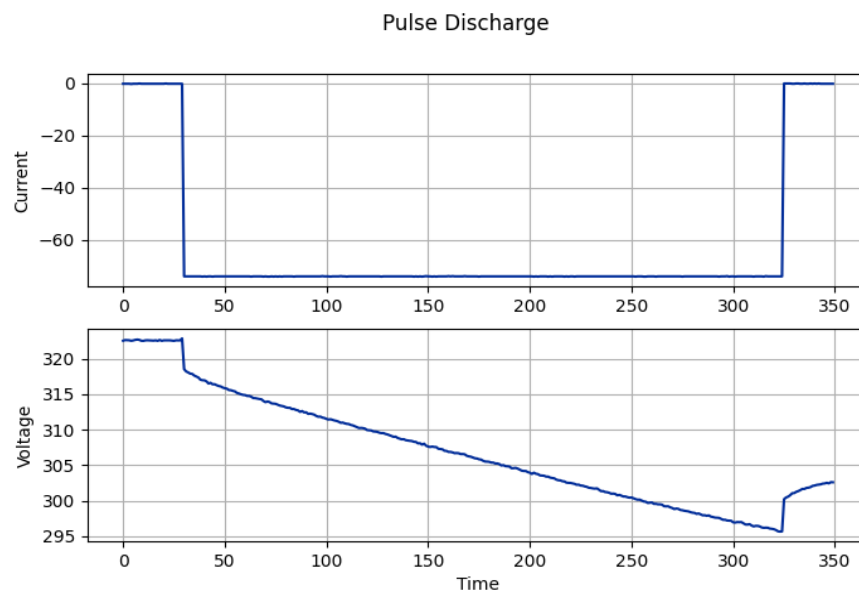
To assess the overall SOH of the battery, a combination of both DCIR and Remaining Capacity is used. These parameters are closely correlated with battery aging, and changes in either or both can indicate a decline in health. The following approach can be used to evaluate SOH:

1. The SOH can be derived by normalizing the DCIR and Remaining Capacity values to their initial (new battery) conditions.
 - $\text{SOH} = (\text{Remaining Capacity} / \text{Rated Capacity}) * 100$ - This is used since rated IR is unknown.
 - $\text{SOH} = (\text{Remaining DCIR} / \text{Rated IR}) * 100$
2. A typical formula for SOH with combined parameters is: $\text{SOH} = \alpha * (\text{Original Capacity} / \text{Remaining Capacity}) + \beta * (\text{Current DCIR} / \text{Initial DCIR})$ Where, α and β are weighing factors typically based on battery type.

Sample Battery Data

The following section provides sample calculations of battery SOH based on resistance, voltage, current, and rated capacity parameters. Figures A-1 and A-2 show sample pulse discharge profiles used to measure internal resistance and remaining capacity, which are key parameters for calculating SOH.

Figure A-1: Battery 1 Pulse Discharge Profile



Source: UCSD

Calculations:

$$\text{DCIR} = (323.62 - 297.85) / (0.21 - (-78.4)) = 32 \text{ mOhms}$$

$$\text{Discharge Capacity} = \text{Integral of current over the discharge period} = 16.11 \text{ Amph}$$

$$\text{Rated Voltage} = 360 \text{ V}$$

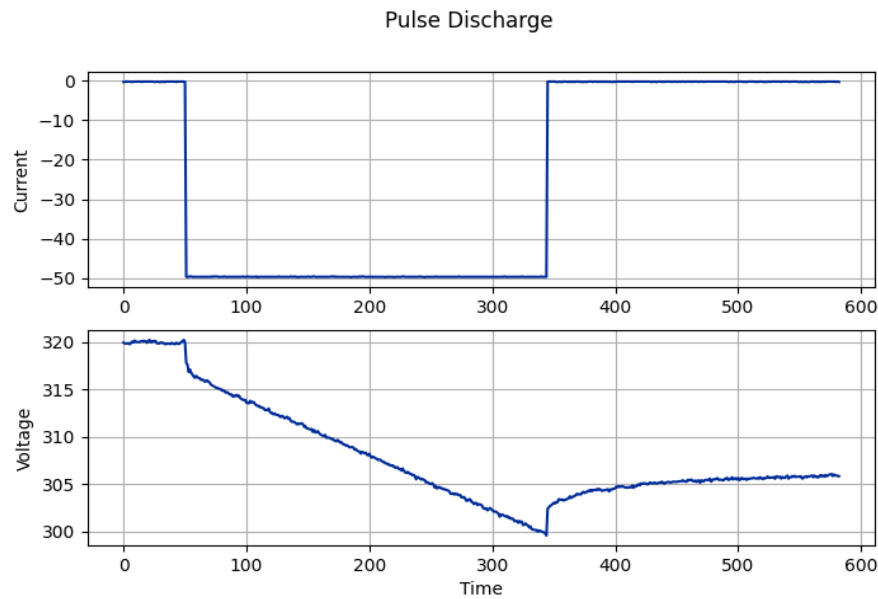
$$\text{Discharge Capacity} = 5.80 \text{ kWh}$$

$$\text{Remaining Capacity} = (5.80 / (0.16 - 0.08)) = 72.54 \text{ kWh}$$

$$\text{Rated Capacity} = 82.1 \text{ kWh}$$

$$\text{SOH} = (72.54 / 82.1) * 100 = 88.36\%$$

Figure A-2: Battery 2 Pulse Discharge Profile



Source: UCSD

Calculations:

$$\text{DCIR} = (320.84 - 302.26) / (0.016 - (-50.03)) = 37 \text{ mOhms}$$

$$\text{Discharge Capacity} = \text{Integral of current over the discharge period} = 11.28 \text{ Amph}$$

$$\text{Rated Voltage} = 360 \text{ V}$$

$$\text{Discharge Capacity} = 4.06 \text{ kWh}$$

$$\text{Remaining Capacity} = (4.06 / (0.23 - 0.17)) = 67.67 \text{ kWh}$$

$$\text{Rated Capacity} = 82.1 \text{ kWh}$$

$$\text{SOH} = (67.67 / 82.1) * 100 = 82.42\%$$

Lithium-Ion Battery Discharging Protocol

To ensure a safe and controlled de-energization process, a two-step discharging protocol is implemented:

1. High-Rate Pulse Discharge: An initial high-rate pulse discharge, typically at a rate of 0.5C to 1C, is applied to rapidly reduce the battery's Voltage to a safe limit. This

rapid discharge accelerates the de-energization process and reduces the risk of thermal runaway.

2. Low-Rate Resistive Discharge: Following the pulse discharge, a resistive discharge is initiated. By connecting a resistor to the battery terminals, the voltage is gradually reduced to a safe level for recycling. This controlled discharge prevents excessive heat generation and ensures a safe and environmentally friendly process.

The following protocol is used for handling a charged cylindrical cell as an example:

1. Connect the cell to the Arbin Tester channel:
 - Attach the positive (red) lead to the positive terminal and the negative (black) lead to the negative terminal.
 - Use the channel's alligator clips to securely attach the cables to the cell terminals.
2. Run a CCCV discharge test:
 - Allow the system to rest for 5 seconds.
 - Perform a constant current discharge at 5A until the cell voltage reaches 2.5V.
 - Switch to constant voltage discharge until the current drops to 0.1A.
 - Allow the system to rest for 5 seconds.
3. Connect the cell to two paralleled 5-ohm resistors using alligator clips (Figure A1):
 - Ensure that the connections are secure for proper current flow.
4. Monitor the cell voltage:
 - Use a multimeter to monitor the voltage until it reaches 0.05V or lower.
5. The cell is now ready for shredding or manual disassembly.

Figure A-3: Instrument and Setups used for Diagnosis and De-energizing



Instruments used for diagnosis

De-energizing batteries

Source: Smartville

Froth Flotation Protocol for Graphite and LCO Separation

Material: Black mass from LCO battery, Deionized (DI) water, Kerosene, and Methyl isobutyl carbinol.

Equipment: Froth Flotation cell and Convection oven. autoclave reactor, magnetic stirrer, vacuum filtration setup, convection oven.

Follow the steps below:-

1. Slurry Formation in Flotation Cell:
 - In a flotation cell, add the LCO black mass and DI water to achieve a solid-to-liquid ratio of 1:5 (e.g., 500g of solids to 2500 mL of liquid), then mix thoroughly to form a slurry.
2. Addition of frothing agent and Kerosene:
 - Add 50 μ L of Methyl isobutyl carbinol to the slurry to act as a frothing agent and 50 μ L of Kerosene as collector, helping to create a stable froth that will carry the graphite particles to the surface.
3. Flotation Process:
 - Start the flotation system and agitate the slurry for 10 minutes to facilitate even mixing.
 - Allow and adjust the airflow for froth formation. The goal is for the graphite particles to attach to the froth and rise to the surface, while the LCO particles remain at the bottom of the cell.
4. Collection and Separation:
 - Skim the froth from the surface, which now contains graphite particles.
 - The remaining slurry in the cell as tail, which contains the LCO could be filtered and dried for the product.

Cathode Active Material Regeneration Protocol (PRIME Process)

Material: NMC111 or NCM622 cathode black mass, 100 g, 4 molar LiOH aqueous solution (300 mL), Deionized (DI) water, Lithium carbonate powder

Equipment: autoclave reactor, magnetic stirrer, vacuum filtration setup, convection oven, box furnace, tube furnace (oxygen atmosphere). Follow the steps below:

1. Hydrothermal Relithiation
 - Add 100 g of NMC111 CBM directly into a 500 mL autoclave reactor containing 300 mL of 4 m LiOH aqueous solution.
 - Seal the reactor and heat at 220 °C for 4 hours.
 - After cooling, gently decant the supernatant and store it for potential reuse.

2. Washing and Filtration

- Add DI water to the remaining solid in the reactor.
- Stir the mixture at 500 rpm for 30 minutes to ensure thorough washing.
- Filter the slurry using vacuum filtration and wash the collected solids with DI water to remove residual lithium salts.

3. Drying

- Transfer the filtered solid to a convection oven and dry overnight at 80 °C.
Note: The yield after drying was 91 wt.% relative to the original CAM weight.

4. Annealing

- Grind the dried material with 5 mol% excess Li_2CO_3 powder to compensate for lithium loss.
- For NMC111, anneal the powder in a box furnace (air atmosphere):
 - Ramp at 5 °C/min to 850 °C, hold for 4 hours, cool naturally to room temperature.
 - For NCM622, follow the same procedure but conduct annealing in a tube furnace under flowing oxygen to maintain oxidation state and crystal structure.