

Recycling of Laptop Spent Li-Ion Batteries and Characterization of Extracted Materials

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As the use of smart devices increases, the energy demand continues to grow, leading to higher consumption of lithium-ion batteries (LIBs) in portable electronics such as laptops, tablets, smartphones, and electric vehicles. This increased usage has resulted in a rising number of discarded batteries, which contain hazardous chemicals and heavy metals that pose serious environmental risks. Recycling these batteries efficiently is essential for both environmental protection and economic sustainability. This study explores a recycling method for used laptop and notebook batteries through a pretreatment and solvent dissolution process, using mild phosphoric acid as the leaching agent. The hydro-metallurgical process successfully recovers 5.124% lithium and 42.143% cobalt, yielding lithium carbonate and cobalt hydroxide. The batteries, which consist of 50.80% lithium cobalt oxide (LiCoO₂) cathodes on aluminum and graphite anodes on copper foils, serve as the primary source of material recovery. The recovered lithium carbonate and cobalt hydroxide are then used to synthesize active powder for cathode material. Advanced characterization techniques, including Cyclic Voltammetry (CV), Raman spectroscopy, and Electrochemical Impedance Spectroscopy (EIS), are employed to analyze the electrochemical properties of the recovered materials and synthesized powders. The results confirm the effectiveness of this recycling method in recovering valuable materials while reducing environmental impact. By addressing the growing problem of battery waste, this approach supports the sustainable production of new batteries through the reuse of critical materials. The study emphasizes the importance of developing efficient recycling technologies to promote a circular economy and reduce dependence on raw material extraction.

Keywords: Lithium-Ion Batteries, Recycling, Hydro-Metallurgy, Lithium Carbonate, Cobalt Hydroxide



Introduction:

In today's rapidly advancing technological world, the demand for high-performance, lightweight, and energy-efficient devices is steadily increasing [1]. Lithium-ion batteries (LIBs), introduced by Sony in 1990, have become essential for powering various devices, from smartphones to electric vehicles, due to their compact size and high energy density [2][3]. However, the limited lifespan of consumer electronics (typically 1 to 3 years) has resulted in a growing number of discarded batteries, raising environmental concerns. Improper disposal of LIBs, which contain toxic substances such as lithium, cobalt, and nickel, poses serious risks to both the environment and valuable resources [4]. Therefore, developing efficient and sustainable recycling methods is critical [5]. Efforts to reduce cobalt usage in LIBs to cut costs have affected battery performance. Cobalt plays a crucial role in improving thermal stability, particle structure, and overall battery capacity. Its reduction or absence presents challenges not only for LIB performance but also for recycling effectiveness [6]. This trade-off highlights the difficulty of balancing cost, efficiency, and recyclability.

Although recycling has clear benefits, traditional methods like pyrometallurgy and the direct method raise significant environmental issues [7]. As the recycling industry expands, these conventional techniques may worsen environmental problems unless more energy-efficient alternatives are developed. To manage varying input materials, impurities, geometries, and changing market needs, adaptable recycling processes are necessary [8]. Additionally, the handling of LIB components requires scalable, standardized, and straightforward processes to ensure efficiency and safety [9]. Hydrometallurgical recycling, though not yet widely commercialized, shows great promise as a future solution for sustainable LIB management. This method enhances the recovery efficiency of cobalt (Co) and lithium (Li) due to the use of hydrogen peroxide as a reducing agent. Hydrogen peroxide promotes the formation of carbonate ions, which have higher solubility in acidic conditions, leading to increased metal extraction during the leaching process. The preferential generation of these easily dissolvable carbonate ions explains the observed improvements in Co and Li recovery rates [10].

The recycling of end-of-life LIBs is a rapidly growing industry with immense potential for future expansion. By prioritizing efficient collection and reuse systems, industries can secure a sustainable supply of critical raw materials while reducing dependence on external sources. Transitioning from informal disposal to formal, systematic LIB recovery frameworks is essential. However, low collection rates, particularly for consumer electronics, remain a key obstacle to scaling up LIB waste processing. Currently, data shows that Asian countries lead in collection efficiency, achieving rates of around 70%, mainly due to their dominance in LIB manufacturing, which incentivizes localized recycling infrastructure [11]. Meanwhile, regions like the European Union, Australia, and the Americas are making gradual progress but often struggle to gather sufficient volumes of spent LIBs to maintain economically viable recycling operations [12].

Handling LIBs requires strict safety protocols, as they are classified as hazardous due to their flammable liquid electrolytes. Potential risks include thermal runaway, which can be triggered by physical damage, extreme heat, or residual electrical charge, leading to fires or explosions during storage, transport, or processing. For instance, crushing discarded LIBs may puncture their casings, while inadequate discharge procedures increase the risk of short circuits. To address these challenges, three primary strategies are being explored for managing LIB waste: refurbishing (repairing for reuse), repurposing (adapting for less demanding applications, such as energy storage), and recycling (recovering raw materials) [13][14].

Novelty Statement:

This study introduces a novel recycling method for spent lithium-ion batteries (LIBs) using mild phosphoric acid as a leaching agent, an approach that has not been widely explored in previous research. Unlike conventional methods that rely on harsh chemicals or energy-intensive processes, this technique is more eco-friendly and energy-efficient. It achieves

impressive recovery rates of 5.124% lithium and 42.143% cobalt, which surpass many existing methods. Another distinctive feature is the synthesis of active cathode material (LiCoO_2) directly from recovered lithium carbonate and cobalt oxide, demonstrating a closed-loop recycling process rarely reported in the literature.

Objectives:

This study aims to tackle the environmental and economic challenges caused by the disposal of spent lithium-ion batteries (LIBs). The goal is to develop an efficient, scalable recycling process that recovers valuable materials like lithium and cobalt, essential for battery production. By using mild phosphoric acid as a leaching agent, the process minimizes environmental impact and reduces energy consumption. Additionally, the study focuses on synthesizing active cathode material (LiCoO_2) from the recovered components, proving the feasibility of a closed-loop recycling system. This approach not only reduces dependence on raw material extraction but also supports a circular economy by reusing critical resources. Ultimately, the research aims to offer a sustainable solution for managing battery waste while addressing the growing demand for energy storage technologies.

This paper highlights the increasing importance of recycling and explores methods to enhance it. It starts by stressing the urgency of recycling and the shift from relying on primary raw materials, like those from conventional mining, to recovering secondary resources from spent lithium-ion batteries (LIBs). The paper will also suggest ways to improve the efficiency and purity of material recovery. The main objective is to recycle lithium and cobalt as Li_2CO_3 and $\text{Co}(\text{OH})_2$ from spent batteries.

Materials and Methods:

For this study, spent lithium-ion batteries (LIBs) were collected from the COMSATS hardware lab. Batteries from HP and Dell laptops were chosen for material extraction and analysis.

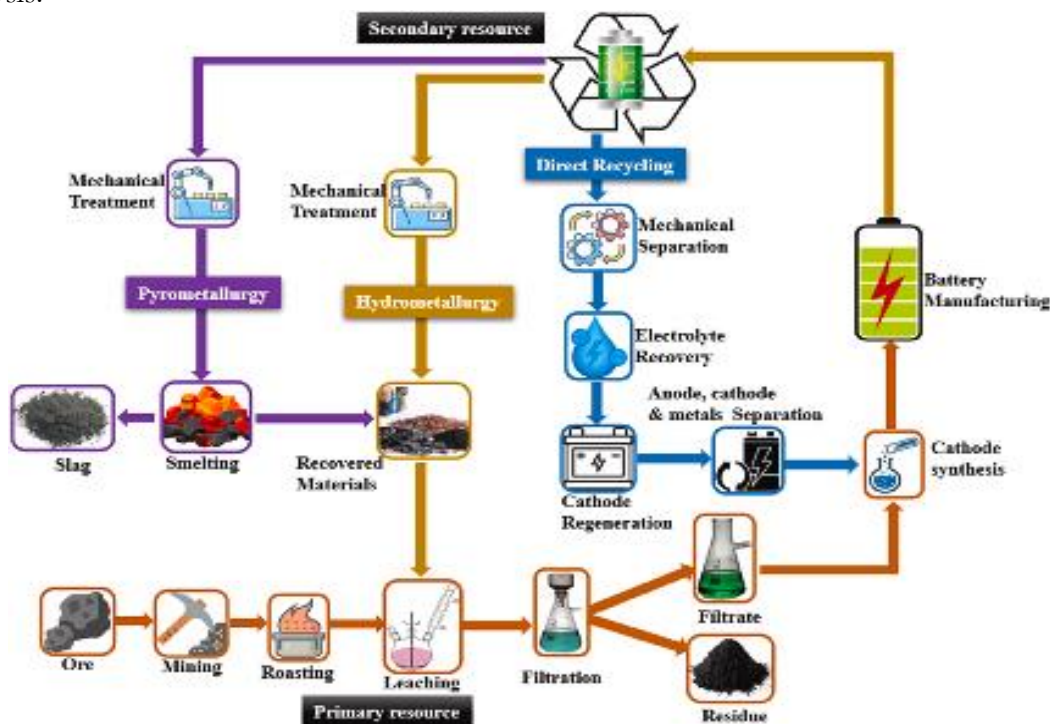


Figure 1. Flow diagram of various lithium-ion battery (LIB) recycling processes

The spent HP notebook battery, consisting of three cells, was selected for analysis. As shown in Table 1, the cathode material in this battery is lithium cobalt oxide (LCO), while the anode is made of graphite. This combination is commonly used in high-performance consumer electronics, such as mobile phones and laptops, due to its balance between energy density and

efficiency. Similarly, the spent Dell laptop battery, containing six cells, was also obtained from the COMSATS hardware lab. According to Table 1, this battery also uses lithium cobalt oxide as the cathode material, while the anode features a thin graphite film applied to the copper foil. This configuration, widely found in portable electronic devices, supports reliable energy delivery and extended cycle life.



Figure 2. Schematic Diagram of Hydrometallurgical recycling procedure

Table 1. Spent HP Notebook and Dell Laptop Battery Specifications

LIB	Cathode Material	Specific Energy (Capacity) Wh/kg	Operating Range (Voltage) V/cell	Cyclic Life	Applications
HP (3 Cells)	LCO	150-200	3.0-4.2	500-1000	Mobile Phones, Laptops, Cameras
Dell (6 Cells)	LCO	170-200	3.0-4.2	600-1500	Mobile Phones, Laptops, Cameras

In addition to the specific batteries mentioned earlier, we also obtained lithium-ion battery (LIB) chemistries, including lithium nickel cobalt manganese oxide (NCM) and lithium manganese oxide (LMO). These chemistries offer distinct advantages based on their metal composition and are commonly used in applications that demand high power and energy density. Table 2 provides a detailed breakdown of the chemical compositions of the LIB chemistries analyzed in this study. Understanding the differences in the relative content of lithium (Li), manganese (Mn), cobalt (Co), and nickel (Ni) is essential for evaluating their electrochemical performance and overall stability.

The recycling process begins by discharging the battery cells to safely eliminate any residual charge. Once discharged, the next step is dismantling, which is performed carefully to avoid damaging the internal components. This process exposes the individual layers of the battery, which are then subjected to various chemical treatments to recover valuable materials. The recovered materials are further processed through controlled methods to ensure optimal purity and efficiency.

Table 2. Chemical Composition of Obtained Different LIBs Chemistries (%)

Chemistry	Li	Mn	Co	Ni
LCO	60.37	0.23	54.43	0.22
LMO	3.85	47.03	0.13	0.02
NMC	6.93	19.28	19.33	20.8

Additional steps in the recycling process include filtration and sieving. The next crucial phase involves leaching, followed by further filtration, to separate valuable elements such as lithium, cobalt, and nickel from impurities. At this stage, the materials are divided into two outputs: sediment and filtrate. The filtrate, which contains the recoverable valuable elements, undergoes a recovery process for purification and refinement. Finally, the recovered materials are analyzed using characterization techniques to assess their quality and confirm whether the desired outcomes have been achieved. These steps not only facilitate the extraction of valuable components but also ensure that the recycling process is both efficient and environmentally sustainable.

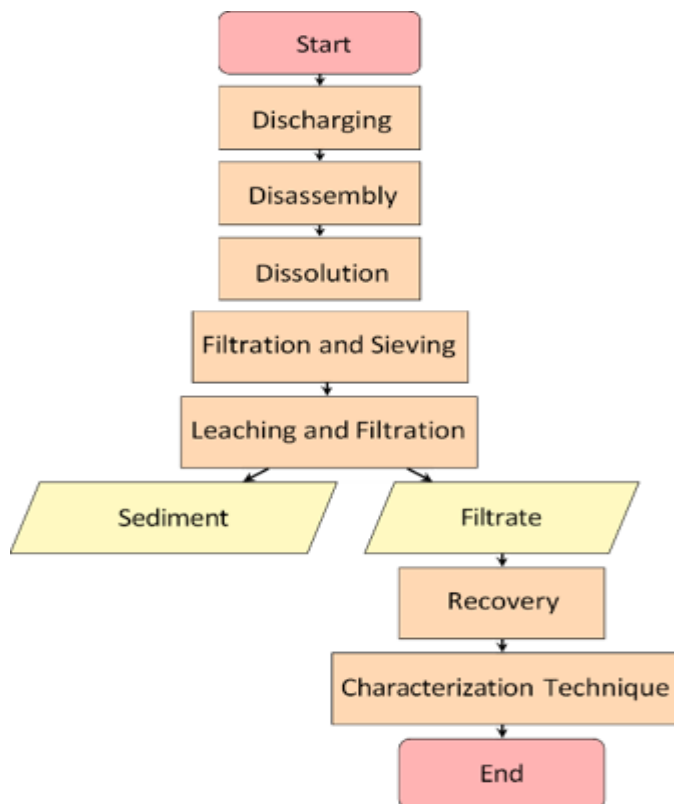


Figure 3. Flow diagram of a methodology for the Recycling Process for Spent LIBs

The recycling process for spent lithium-ion batteries (LIBs) begins with the crucial step of discharging the batteries to eliminate any remaining charge. Even after use, these batteries may retain 2-5% of their charge, which, if not properly discharged, could lead to short-circuiting and serious hazards such as explosions. To prevent this risk, the batteries were fully discharged before further processing.

The discharge process involved submerging the battery cells in a salt solution containing 500 mL of deionized water and 5 wt% sodium chloride (NaCl) for 24 hours inside a fume hood. During this period, gases such as nitrogen (N_2), carbon dioxide (CO_2), water vapor (H_2O), hydrocarbons (C_xH_y), and acetic acid ester (CH_3COOCH_3) were released. This process separates the supernatant, which contains small amounts of lithium (Li), cobalt (Co), and phosphorus (P), from the sediment, which includes traces of aluminum (Al) and iron (Fe).

By fully discharging the batteries, the risk posed by residual charge is eliminated, making them safe for the subsequent stages of recycling and material recovery.



Figure 4. Discharging notebook battery cell

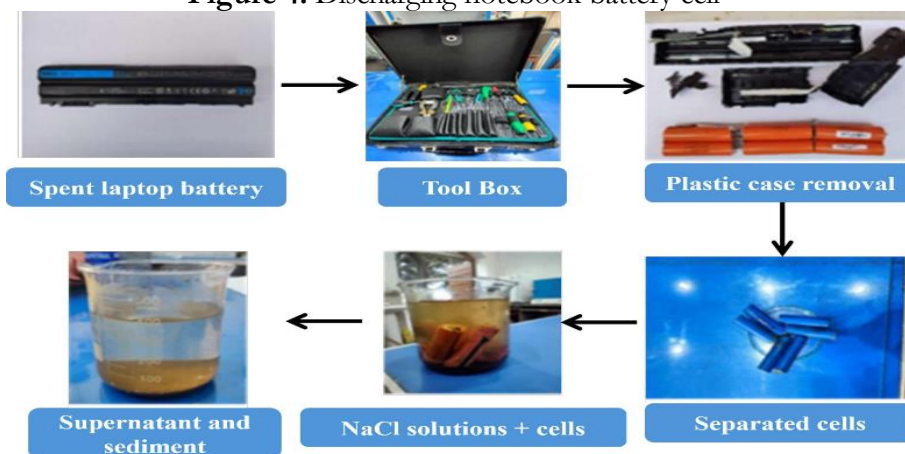


Figure 5. Discharging process of laptop battery cell

After discharging, the batteries were carefully dismantled. Using pliers and a blade, the plastic casing and electrical circuits were removed to expose the rectangular cells from the notebook batteries (Figure 4) and the cylindrical cells from the laptop batteries (Figure 5). Once separated, the cells were placed in an oven and dried at 60°C for 12 hours to ensure they were completely free of moisture before further dismantling.

Next, the dried cells were processed carefully by cutting and peeling off the plastic and metal shields. To safely open the metal casings, both ends were cut using a lathe machine, while a milling machine was employed to make precise incisions in the cylindrical metal shells. Once opened, the layers of the anode, separator, and cathode were carefully unrolled and separated, preparing them for subsequent processing and material recovery.

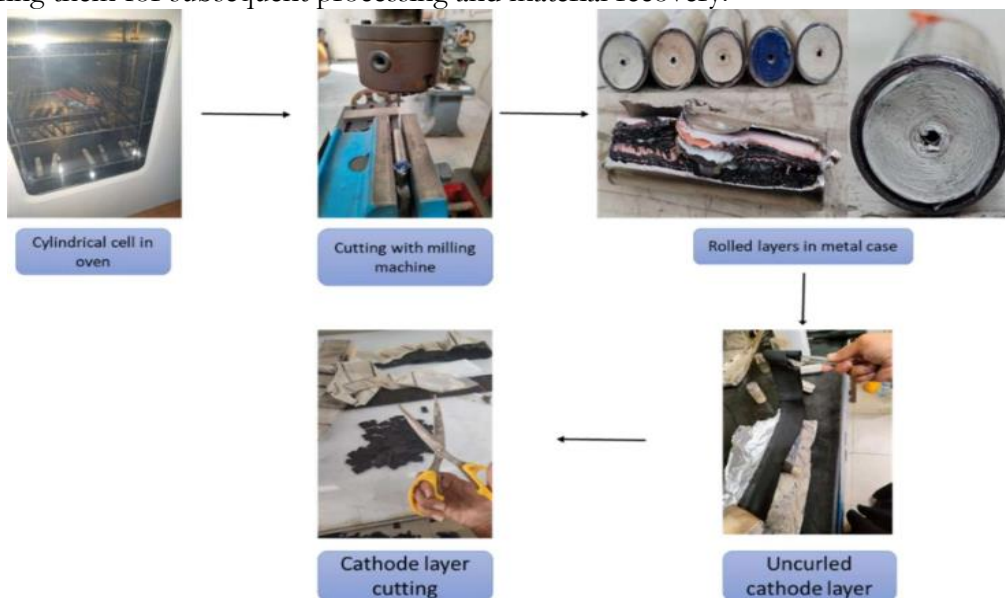


Figure 6. Dismantling process of the battery cells

The next step in the process involved the solvent dissolution method as shown in figure 7, which aimed to weaken the adhesion of the cathode material to the aluminum foil, making extraction easier. The binders, typically polyvinylidene fluoride (PVDF) or polytetrafluoroethylene (PTFE), posed challenges during separation. To address this, an organic solvent, N-Methyl-2-pyrrolidone (NMP), was used to dissolve the binders.

The aluminum foil was first cut into smaller pieces and then immersed in NMP at 130°C for 24 hours in an oven placed inside a fume hood. During this process, the cathode material is mixed with the solution, leaving the aluminum foil intact. The cathode material was subsequently recovered through filtration, followed by drying and grinding the filtrate residue to obtain a fine cathode powder.

This powder was then subjected to leaching to extract valuable metals such as lithium and cobalt. Phosphoric acid (0.7 moles) was used as the leaching agent and placed in a beaker on a magnetic stirrer set to a constant temperature of 40°C. Once the solution reached the desired temperature, the powdered cathode material was added, and hydrogen peroxide (H_2O_2) was added dropwise to enhance the leaching efficiency. The mixture was stirred continuously for one hour as shown in figure 8 and then allowed to cool to room temperature.

The leaching process successfully extracted lithium and cobalt ions from the cathode material into the solution. The resulting liquid was filtered to separate the sediment, which contained residual lithium, cobalt, carbon, and other impurities. The filtrate was then processed further to recover purified lithium and cobalt.

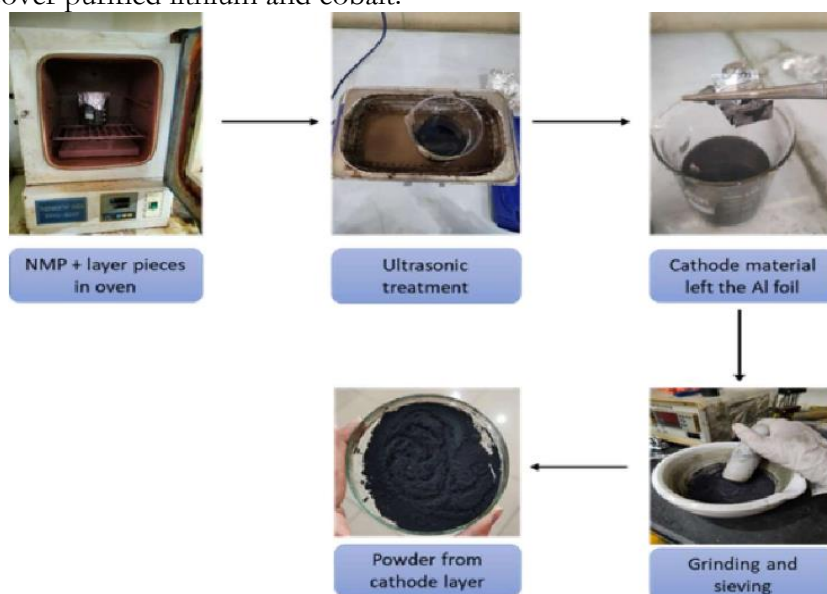


Figure 7. Cathode powder detached from aluminum foil

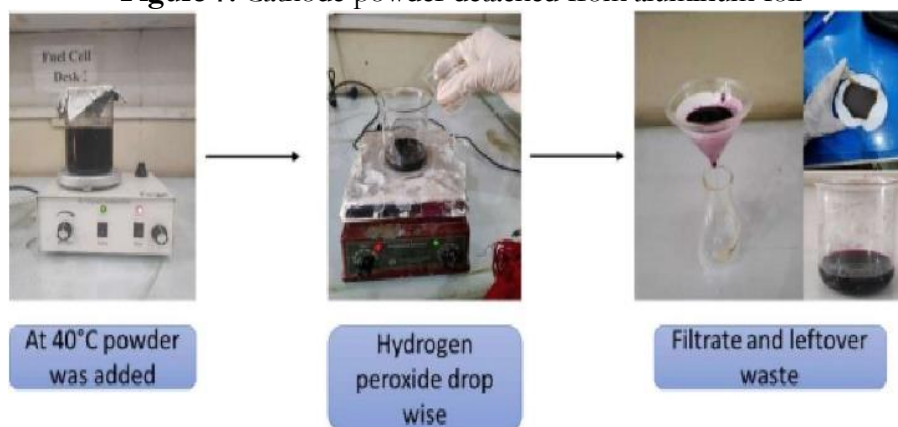


Figure 8. Leaching of filtrate

To recover cobalt from the filtrate, the solution was placed on a magnetic stirrer, and a 2M sodium hydroxide (NaOH) solution was added drop by drop, acting as a reducing agent. The pH level was continuously monitored using a pH meter figure 9 throughout the process.

As the pH reached 6, cobalt began to precipitate, forming violet-colored particles. This precipitation continued until the pH level rose to 8. The resulting mixture was then filtered to collect the violet cobalt precipitates on filter paper. These precipitates were dried to obtain cobalt hydroxide in powdered form, which served as a key intermediate product for further processing.

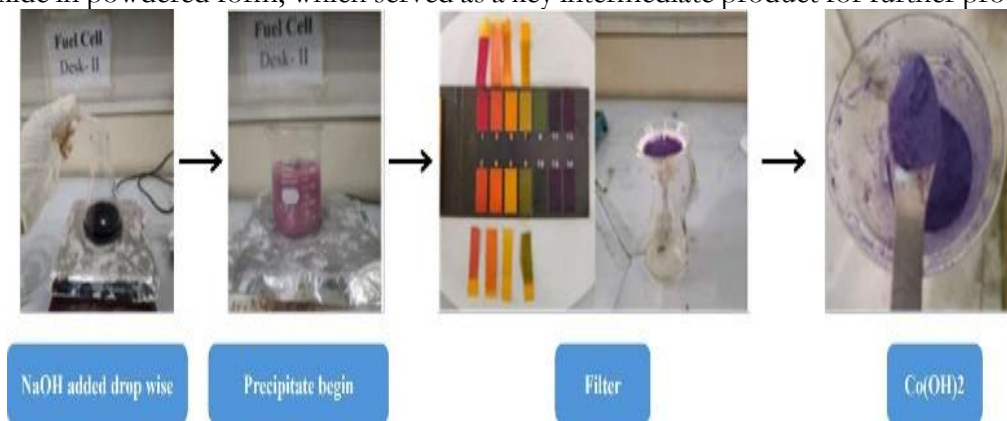


Figure 9. Recovery of Cobalt

To convert cobalt hydroxide into cobalt oxide, hydrogen peroxide (H_2O_2) was added dropwise to the cobalt hydroxide solution under constant stirring (Figure 10). This reaction led to the formation of brown cobalt oxide precipitates.

The solution was then placed in an ultrasonic bath set to $50^\circ C$ and left for 5 hours, which helped refine and enhance the quality of the cobalt oxide precipitates. Afterward, the mixture was filtered, and the collected precipitates were dried to obtain cobalt oxide in its final powdered form.

The resulting filtrate from this process was subsequently used for lithium recovery.

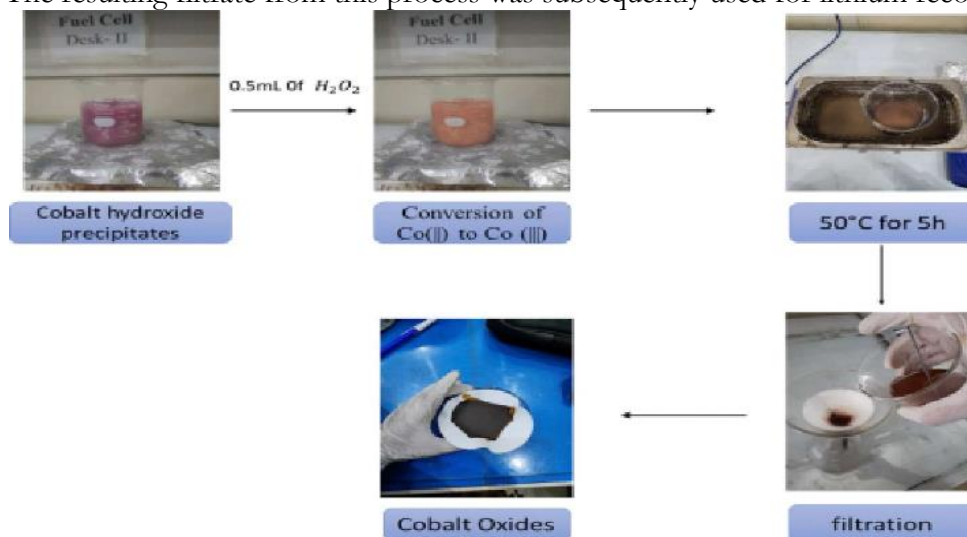


Figure 10. Brown precipitates of cobalt oxide from cobalt hydroxide

For lithium recovery, the filtrate was heated to $100^\circ C$ while being continuously stirred on a magnetic stirrer. Sodium carbonate (Na_2CO_3) solution was then added drop by drop until white lithium carbonate precipitates began to form, as shown in Figure 11.

The precipitates were subsequently filtered and dried to obtain lithium carbonate in powdered form.

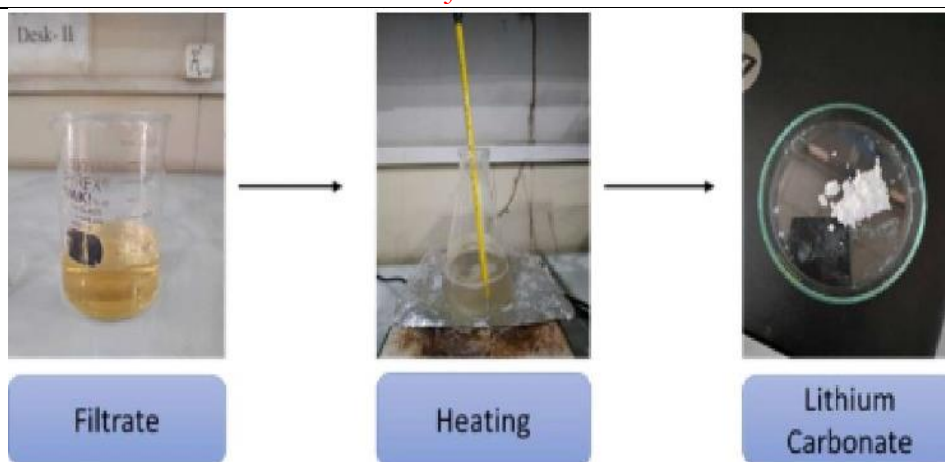


Figure 11. Recovery of lithium

Preparation of Active Material for Cathode:

Synthesis of Lithium Cobalt Oxide (LCO):

Figure 12 shows the flow chart for synthesizing the active powder. The extracted lithium carbonate and cobalt oxide particles were centrifuged to remove any remaining impurities. These recovered materials, obtained from spent LIBs, were then reused to prepare the cathode active material.

The purified cobalt oxide and lithium carbonate were thoroughly mixed and ground in a mortar before being calcined at 800°C for approximately 12 hours in a muffle furnace. After calcination, the resulting powder was re-grounded and sintered at 850°C to enhance its properties. This process yielded lithium cobalt oxide (LCO) powder, which was ready for use as cathode material.

Data on Recovered Material:

The recovered materials from the laptop and notebook batteries, along with their respective weights (in grams), are summarized in Table 3 below.

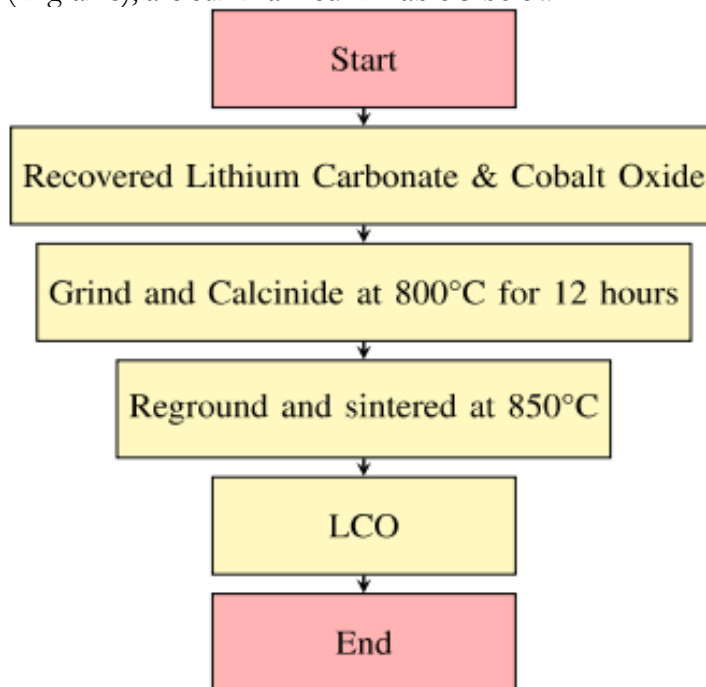


Figure 12. Flowchart for synthesis of Active Powder.

Table 3. Recovered Materials Data

Laptop Battery	Weight (g)	Note Book Battery	Weight (g)
Cathode Powder	20g	Cathode Powder	10g
Anode Powder	15g	Anode Powder	18g
Aluminium Foil	10g	Aluminium Foil	07g
Copper Foil	10g	Copper Foil	08g
Plastic and Separators	18g	Plastic and Separators	14g
Metal Case	25g	Metal Case	20g
Cobalt Hydroxide, Lithium Carbonate	8g, 4g	Cobalt Hydroxide, Lithium Carbinat	5g, 3g

Result and Discussion:

Raman Spectroscopy:

The Raman spectroscopy analysis of lithium carbonate, shown in Figure 13a, reveals characteristic peaks associated with the vibrational modes of both the carbonate ion (CO_3^{2-}) and lithium ions. The prominent band at approximately 1088 cm^{-1} corresponds to the symmetric stretching vibrations of the CO_3^{2-} ion, reflecting its planar structure. Weaker bands at around 748 cm^{-1} and 712 cm^{-1} represent in-plane bending vibrations, while a smaller band at $\sim 1458\text{ cm}^{-1}$ is attributed to the asymmetric stretching of the C–O bonds. These peaks confirm the identity of lithium carbonate (Li_2CO_3) and provide crucial insights into its bonding, symmetry, and structural properties. This makes Raman spectroscopy an invaluable tool for investigating the role of lithium carbonate in batteries and energy storage technologies.

In Figure 13b, the Raman spectrum of cobalt hydroxide ($\text{Co}(\text{OH})_2$) displays distinct peaks at 420 cm^{-1} and 510 cm^{-1} , which correspond to O–H bending vibrations and the Co–O symmetric stretching mode (Ag), respectively. These sharp, well-defined peaks indicate the material's high crystallinity and provide essential information about the bonding environment of cobalt and hydroxide ions. This structural characterization highlights the potential of cobalt hydroxide for applications in energy storage and catalysis.

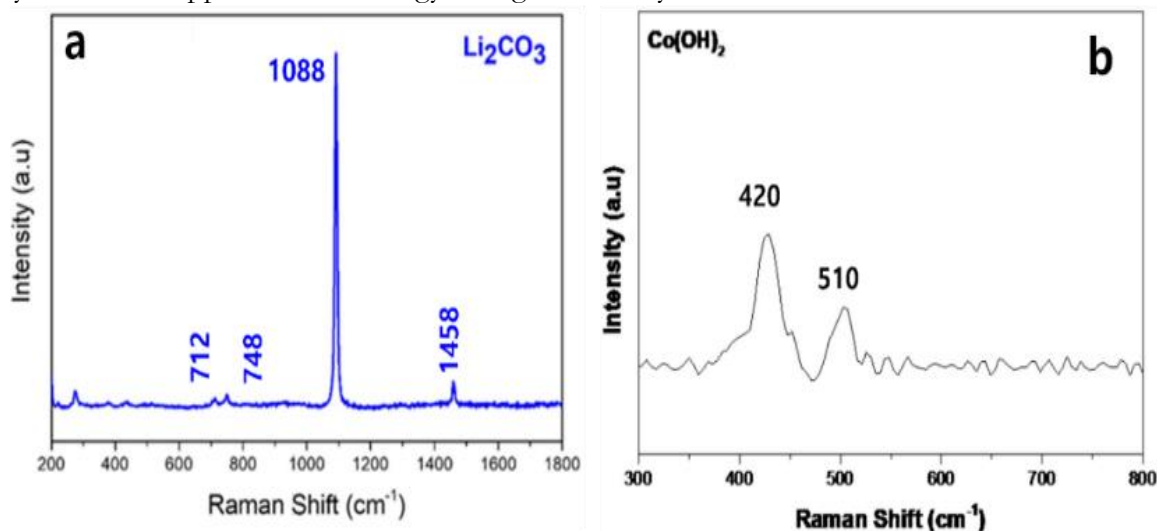


Figure 13. Raman Spectroscopy of (a) Lithium Carbonate (Li_2CO_3) (b) Cobalt Hydroxide ($\text{Co}(\text{OH})_2$)

X-ray Diffraction:

XRD Analysis of Lithium Carbonate and Cobalt Hydroxide:

The XRD pattern of lithium carbonate (Li_2CO_3), shown in Figure 14a, confirms its monoclinic crystal structure with a space group of $C2/c$. Prominent diffraction peaks are

observed at approximately 21.4° , 30.6° , 36.0° , 39.5° , and 43.8° (2θ), corresponding to characteristic lattice planes such as (110) and (211). These sharp peaks reflect the high crystallinity and phase purity of the sample, with minimal structural defects. This well-crystallized nature enhances the thermal stability and electrochemical performance of lithium carbonate, making it highly suitable for applications that require structural integrity and reliability, such as battery technologies or as a precursor for other lithium-based compounds. Furthermore, the precise alignment of lattice planes indicates that the synthesis method employed was effective in achieving the desired phase with minimal impurities or defects.

The XRD pattern of cobalt hydroxide ($\text{Co}(\text{OH})_2$), shown in Figure 14b, exhibits a prominent low-angle diffraction peak at 2θ corresponding to the (003) basal reflection, which represents the interlayer d -spacing. This peak is characteristic of the layered double hydroxide (LDH) structure of $\text{Co}(\text{OH})_2$. However, due to the intercalation of anions (e.g., NO_3^- , Cl^- , CO_3^{2-}) and water molecules within the layers, $\text{Co}(\text{OH})_2$ exhibits a more disordered layered structure, resulting in broader and less defined XRD peaks. This irregular alignment of layers reduces the overall crystallinity of the material. Higher-order reflections, such as (006) and (009), may appear at regular intervals due to the periodic stacking of the layers. The exact position and intensity of the peaks may vary depending on factors such as the type of intercalated anions and the degree of hydration or water content in the structure. These structural characteristics influence the material's potential applications in catalysis, energy storage, and electrochemical processes.

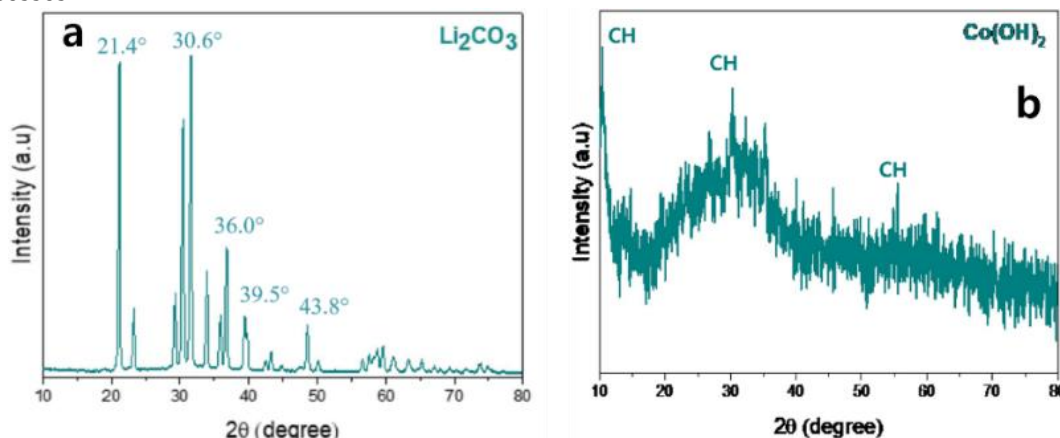


Figure 14. XRD pattern of (a) Li_2CO_3 showing distinct peaks indicative of its crystalline structure (b) $\text{Co}(\text{OH})_2$ showing distinct diffraction peaks that suggest the structure and phase purity of the sample.

Fourier Transform Infrared Spectroscopy (FTIR):

The FTIR spectrum of lithium carbonate (Li_2CO_3) displays key peaks that confirm its structure. The peak at 1412 cm^{-1} corresponds to the asymmetric stretching vibrations of the carbonate ion, a crucial feature that confirms the presence of carbonate groups. The peak at 1087 cm^{-1} represents the symmetric stretching mode of the carbonate ion, indicating well-defined carbonate bonding. The peak at 859 cm^{-1} is attributed to the out-of-plane bending vibrations of the carbonate ion, reflecting the structural stability of Li_2CO_3 . Similarly, the peak at 739 cm^{-1} is associated with the bending vibrations of the carbonate ion, further validating the vibrational properties of lithium carbonate.

The absence of additional peaks, such as those linked to hydroxyl groups or impurities, indicates the high purity of the sample. The observed spectral features match previously reported FTIR data for lithium carbonate, confirming its structural identity.

In addition, the peak at 1054 cm^{-1} in the FTIR spectrum is characteristic of vibrational modes in cobalt-based materials, representing Co–OH bonds. Peaks at 1374 cm^{-1} and 1636

cm^{-1} arise from the intercalation of OH^- and NO_3^- anions, respectively. This ion intercalation is a distinctive feature of $\text{Co}(\text{OH})_2$, enhancing mass and electron transport between the catalyst and the electrolyte during electrocatalysis.

The peak at 3494 cm^{-1} corresponds to O–H stretching, which is characteristic of hydroxyl groups or water molecules. This suggests that the material is either hydrated or contains surface-bound water, as observed in $\text{Co}(\text{OH})_2$. These peaks imply that $\text{Co}(\text{OH})_2$ or its hydrated form may influence electrochemical properties by modifying surface chemistry and ion transport, which, in turn, affects energy storage performance in applications such as supercapacitors or batteries.

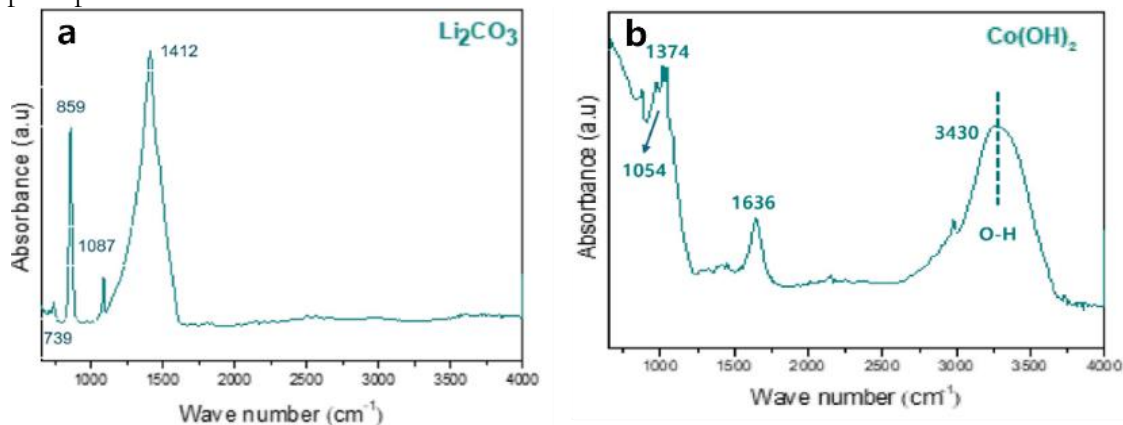


Figure 15. FTIR pattern of (a) Li_2CO_3 showing distinct peaks indicative of its crystalline structure and (b) $\text{Co}(\text{OH})_2$ showing distinct diffraction peaks that suggest the crystalline structure and phase purity of the sample.

Electrochemical Impedance Spectroscopy (EIS):

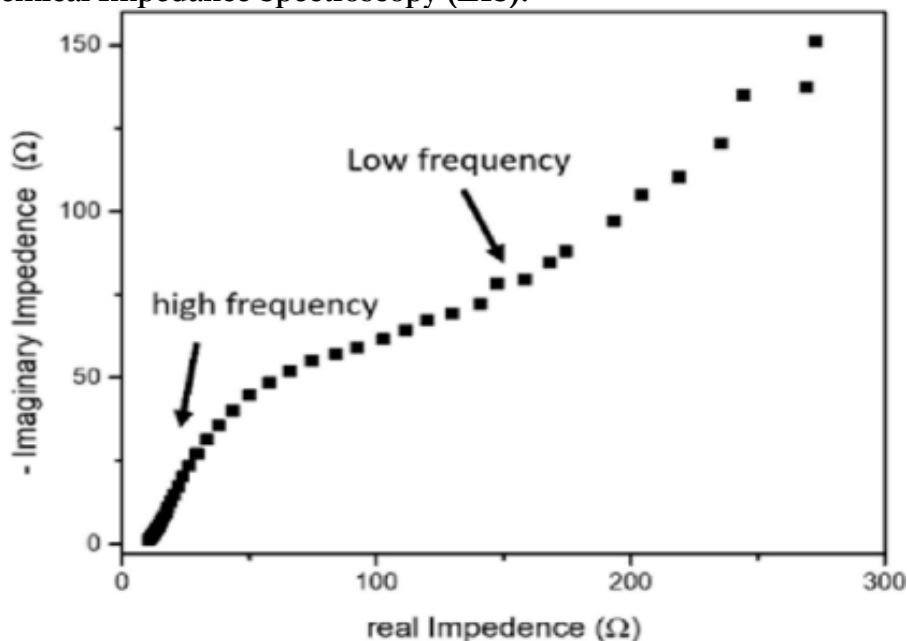


Figure 16. Nyquist plot of Cobalt oxide from EIS.

Nyquist EIS Curve:

The Nyquist EIS curve of cobalt oxide (Co_xO_y) shows a half-semicircle at high frequencies, indicating low charge transfer resistance (R_{ct}). This suggests efficient electron transfer at the electrode-electrolyte interface. Following the semicircle, the curve transitions into an upward diagonal line, which represents Warburg impedance, reflecting ion diffusion into the

material. This indicates a combination of capacitive and diffusion-controlled charge storage mechanisms.

The half-semicircle implies fast charge transfer kinetics, while the diagonal line indicates moderate ion diffusion limitations, typical of pseudocapacitive materials. Overall, this behavior demonstrates that cobalt oxide has high-rate capability, making it suitable for high-power energy storage applications, such as supercapacitors or hybrid capacitors, with good cycling stability and efficient electrochemical performance.

Cyclic Voltammetry (CV):

The cyclic voltammetry (CV) of cobalt oxide (Co_xO_y) exhibits a rectangular shape, which is characteristic of pseudocapacitive behavior. In this case, charge storage occurs mainly through surface-controlled redox reactions, unlike the intercalation process observed in traditional battery materials. The absence of well-defined redox peaks suggests that the electrochemical reactions are fast and reversible, enabling rapid charge and discharge cycles.

This behavior is typical of materials that combine both capacitive and faradaic charge storage mechanisms, often seen in transition metal oxides like cobalt oxide. These materials can store charge electrostatically and through surface redox processes. The rectangular CV shape in Fig. 17a reflects high-rate capability and fast electron transfer, making cobalt oxide ideal for high-power applications such as supercapacitors or hybrid energy storage systems.

Additionally, the observed behavior indicates that cobalt oxide offers good cycling stability and long-term performance. The stable electrochemical properties suggest that recovered cobalt oxide can be effectively reused in subsequent cycles, providing a sustainable and cost-effective option for future energy storage technologies.

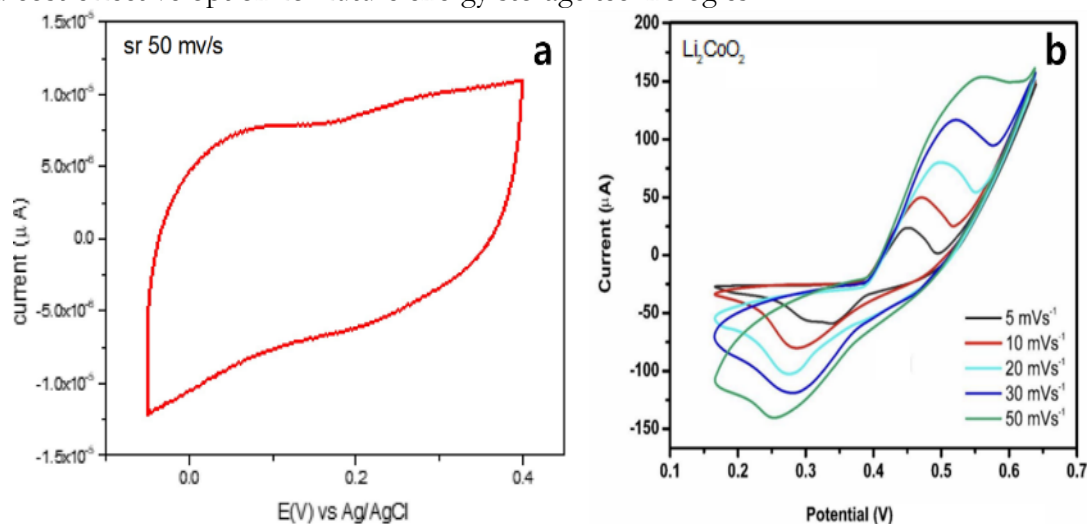


Figure 17. Cyclic Voltammetry of (a) Cobalt Oxide and (b) LiCoO_2 (LCO) active material

The cyclic voltammetry (CV) analysis of the recovered LiCoO_2 in Fig. 17b shows a distinctive profile, characterized by pronounced peaks near the potential window edges and a near-convergence of oxidation and reduction peaks in the central region. This convergence in the middle suggests highly reversible redox processes with minimal polarization, indicating efficient charge transfer kinetics. In contrast, the distinct peaks at the edges may reflect the activation of additional electrochemical processes or slight diffusion limitations at extreme potentials.

Overall, these features highlight a complex balance between surface-controlled pseudocapacitive behavior and diffusion-limited kinetics. This CV profile emphasizes the strong electrochemical performance of the recycled LiCoO_2 , demonstrating its suitability for high-rate applications and showcasing the potential of recovered materials in sustainable energy storage technologies.

Discussion:

The results of this study demonstrate the feasibility of recycling lithium-ion batteries (LIBs) from spent laptop cells using a hydrometallurgical process with mild phosphoric acid leaching, achieving significant lithium (5.124%) and cobalt (42.143%) recovery rates. The successful synthesis of LiCoO_2 cathode material from recovered metals highlights the potential of this process in promoting closed-loop LIB recycling, aligning with global sustainability goals for resource conservation and electronic waste management. Compared to conventional sulfuric acid and hydrochloric acid leaching methods, the use of phosphoric acid offers a less corrosive, more environmentally friendly approach, reducing the formation of secondary pollutants and minimizing hazardous waste disposal challenges. These findings contribute to recent advancements in sustainable battery recycling technologies, supporting efforts to replace energy-intensive pyrometallurgical processes with cost-effective hydrometallurgical alternatives (Makuza et al., 2021).

One of the key advantages of this study is the comprehensive characterization of recovered materials, which confirms the structural integrity and electrochemical performance of regenerated LiCoO_2 . XRD and Raman Spectroscopy results indicate that the synthesized cathode material retains a stable layered structure, similar to commercially available LiCoO_2 , while Electrochemical Impedance Spectroscopy (EIS) and Cyclic Voltammetry (CV) confirm its favorable charge-transfer properties. However, while initial results suggest promising electrochemical behavior, long-term cycling stability tests are required to assess capacity retention and degradation mechanisms over extended charge-discharge cycles. Previous studies (Richa et al., 2017) have shown that recycled LiCoO_2 may exhibit slightly lower energy density and cycle life due to impurity incorporation during the recovery process. Future work should focus on optimizing purification steps to enhance material performance and compare the electrochemical stability of recovered LiCoO_2 against commercially synthesized cathodes.

Despite the positive outcomes, certain challenges remain regarding the scalability and economic feasibility of phosphoric acid leaching for large-scale LIB recycling. While phosphoric acid is less hazardous than strong mineral acids, its leaching efficiency for lithium is relatively lower compared to citric acid or oxalic acid-based processes, which have been reported to achieve higher lithium recovery rates with minimal environmental impact (Xie et al., 2021). Additionally, the study does not evaluate the energy consumption of the leaching, precipitation, and calcination steps, which is critical for determining the commercial viability of this method. Future research should incorporate a techno-economic analysis to assess the cost-effectiveness, reagent consumption, and energy demands associated with phosphoric acid leaching, as well as explore hybrid recycling approaches that combine mechanical, hydrometallurgical, and direct cathode regeneration techniques for maximum resource recovery.

Another crucial aspect that requires further exploration is the management of leachate residues and wastewater treatment. While hydrometallurgical methods produce fewer emissions than pyrometallurgy, they still generate waste streams containing dissolved metals, residual acids, and organic binders that require proper treatment before disposal. Previous research (Werner et al., 2020) emphasizes that leaching residues can contribute to secondary pollution if not adequately neutralized or processed for further metal recovery. Future studies should focus on developing closed-loop wastewater treatment systems that enable safe disposal or reuse of leachate solutions to minimize environmental impacts. Additionally, investigating the potential for recovering additional byproducts such as aluminum, copper, and graphite from spent batteries could further improve the sustainability and profitability of LIB recycling.

In conclusion, this study successfully demonstrates that phosphoric acid-based hydrometallurgical recycling is an effective method for lithium and cobalt recovery from spent LIBs, offering a sustainable alternative to conventional battery disposal. However, further optimizations are needed to enhance lithium recovery efficiency, improve the electrochemical

stability of regenerated cathode materials, and address environmental concerns related to wastewater treatment and process scalability. By integrating advanced purification techniques, optimizing reaction conditions, and conducting large-scale feasibility assessments, future research can contribute to the development of commercially viable LIB recycling solutions, supporting the transition toward a circular economy in battery materials management.

Limitations:

Hydrometallurgical recycling of lithium-ion batteries faces challenges related to scalability, cost, and environmental impact. Scalability is limited by complex multi-step processes, including leaching, separation, and purification, which must be tailored to accommodate variations in battery chemistry. High costs arise from the use of expensive reagents (e.g., acids and solvents), energy-intensive operations (such as heating and evaporation), and wastewater treatment to handle toxic effluents containing heavy metals and fluorides.

The recycling process generates chemical waste, including acidic/alkaline wastewater and metal-laden sludge, which requires proper disposal to prevent environmental harm. Technical challenges include lithium losses due to its solubility, difficulties in separating chemically similar metals (e.g., cobalt and nickel), and interference from battery binders (e.g., PVDF). Despite offering precise metal recovery and lower energy consumption compared to pyrometallurgy, these limitations underscore the need for innovative solutions, such as closed-loop recycling systems and bioleaching, to improve the efficiency and sustainability of the process.

Conclusion:

This study successfully developed a scalable and efficient recycling process for spent lithium-ion batteries sourced from laptops and notebooks, using mild phosphoric acid as a leaching agent. The process achieved notable recovery rates, yielding 5.124% lithium and 42.143% cobalt, which were subsequently converted into lithium carbonate and cobalt hydroxide. Comprehensive characterization confirmed the high purity and structural integrity of these recovered materials. Additionally, cobalt oxide extracted during the process was used to produce LCO (lithium cobalt oxide) active powder.

These findings demonstrate the feasibility of repurposing materials from spent batteries, offering a sustainable alternative to traditional waste disposal methods that often harm the environment. The purified lithium carbonate and cobalt hydroxide not only present opportunities for reuse in battery manufacturing but also help address resource scarcity amidst rising demand for lithium-ion batteries.

Ultimately, this research highlights the importance of developing efficient recycling technologies to support a circular economy in battery production. By recovering valuable materials, the approach addresses environmental concerns while enhancing the economic viability of recycling initiatives.

Looking ahead, the outlook for lithium-ion battery (LIB) recycling is promising but requires further innovation and collaboration to overcome existing challenges. Enhancing leach efficiency, reducing energy consumption, and improving recovery rates for valuable metals like lithium and cobalt will be crucial. The development of direct recycling methods that preserve cathode structures could significantly minimize environmental impact and energy use.

Moreover, integrating automation and AI in recycling operations can further streamline the process. Exploring alternative battery chemistries, such as lithium iron phosphate (LFP) or sodium-ion batteries, could also reduce dependence on critical materials. Policymakers have a key role to play by implementing regulations and incentives to promote recycling and circular economic practices.

Global collaboration and standardization of recycling protocols will be essential to address the growing problem of LIB waste on a larger scale. By advancing these areas, the industry can

work toward a more sustainable, resource-efficient future, ensuring that the increasing demand for energy storage solutions is met without compromising environmental sustainability.

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