

Efficient Recycling of Spent LiCoO₂ Cathodes Via Confined Pore-Assisted Simplified Direct Carbothermic Reduction Without External Reducing Agents

Donghun Kang, Joowon Im, Sujong Chae*, Haeseong Jang*, and Minseong Ko* 

As demand for lithium-ion batteries increases, the supply of materials is increasingly constrained by their geographical concentration. This has spurred significant research into recycling spent batteries to enhance resource circulation. Currently, commercially applied recycling methods (such as pyrometallurgy and hydrometallurgy) face environmental and economic challenges, including waste acid and gas generation, high-temperature heat treatment, and operational complexity. A promising alternative is the carbothermic reduction process, which operates at lower temperatures, minimizing costs and environmental emissions. However, this method still requires large quantities of external reducing agents. Therefore, this study aims to introduce a simplified direct carbothermic reduction (SDCR) process. The SDCR process leveraged carbon conductive materials and organic binders within the electrode as reducing agents. Additionally, the high compaction state created a conducive environment for reducing gases, promoting efficient reduction and material recovery. This approach reduces the reliance on external reducing agents and streamlines the re-upcycling process, making it commercially viable.

1. Introduction

The demand for lithium-ion batteries (LIBs), initially used primarily in small IT devices, has surged recently. Due to the commercialization of eco-friendly transportation, such as electric vehicles, as part of global carbon neutrality initiatives.^[1–3] A recent report projects a rapid increase in the demand for LIBs from 100 GW in 2020 to over 6530 GW, more than 60-fold over the next 30 years.^[4] Given their average life span of 5–8 years, the volume of spent LIBs is expected to rise

significantly,^[5–8] posing environmental pollution risks.^[9–12] Moreover, the raw materials required for these batteries are geographically concentrated in regions such as Latin America and Africa, which could create an unstable supply of raw materials.^[12–14] Efforts to effectively recycle raw materials from spent LIBs are actively underway to improve resource circulation.^[15–17]

Different efforts to recycle LIBs focus on recovering valuable metals, particularly Li and Co, from cathode materials.^[18–22] Two primary methods, hydrometallurgy and pyrometallurgy, are commonly employed for this purpose. In hydrometallurgy, recycling relies on strong acids like nitric or sulfuric acid, generating large volumes of wastewater that lead to high treatment costs and environmental pollution.^[23–25] To address these issues, researchers are exploring eco-friendly organic acids, such as deep eutectic solvents.^[26–28] However, these approaches are limited by low leaching rates. In pyrometallurgy, high-temperature treatments

exceeding 1000 °C are used to recover materials, but the process is energy-intensive, increasing recycling costs.^[29–31] To mitigate these challenges, studies are investigating methods, such as carbothermic reduction, which involves adding a reducing agent to reactants at relatively lower temperatures.^[32–34]

Previous studies focus on utilizing reducing agents, such as graphite from anode electrodes or eco-friendly alternatives, such as biomass waste, while optimizing proportions and temperature conditions based on reducing agent type.^[35–37] Furthermore, mixing the reducing agents with spent cathode materials requires an extra step to separate the aluminum current collector. Minimizing the energy consumption of heat treatments in carbothermic reduction demands substantial amounts of reducing agents. Therefore, understanding the reduction mechanism of spent cathode materials is crucial. Developing effective strategies to minimize the reducing agent usage and energy consumption by simplifying the recycling process is essential for improving sustainability.

This study aims to develop a strategy for simplified direct carbothermic reduction (SDCR) that simultaneously peels and reduces the spent cathode material within the electrodes. The materials recycled through this process were upcycled into single-crystal, Ni-rich cathode materials to achieve high-energy-density LIBs with long cycle life. The carbothermic reduction process involves solid–solid and solid–gas

D. Kang, J. Im, Prof. M. Ko
Department of Metallurgical Engineering, Pukyong National University,
Busan 48513, Korea
E-mail: msk0876@pknu.ac.kr

Prof. S. Chae
Division of Applied Chemical Engineering, Pukyong National University,
Busan 48513, Korea
E-mail: schae@pknu.ac.kr

Prof. H. Jang
Department of Advanced Materials Engineering, Chung-Ang University,
Anseong-si, Gyeonggi-do 17546, Korea
E-mail: hjang4765@gmail.com

 The ORCID identification number(s) for the author(s) of this article can be found under <https://doi.org/10.1002/eam2.70037>.

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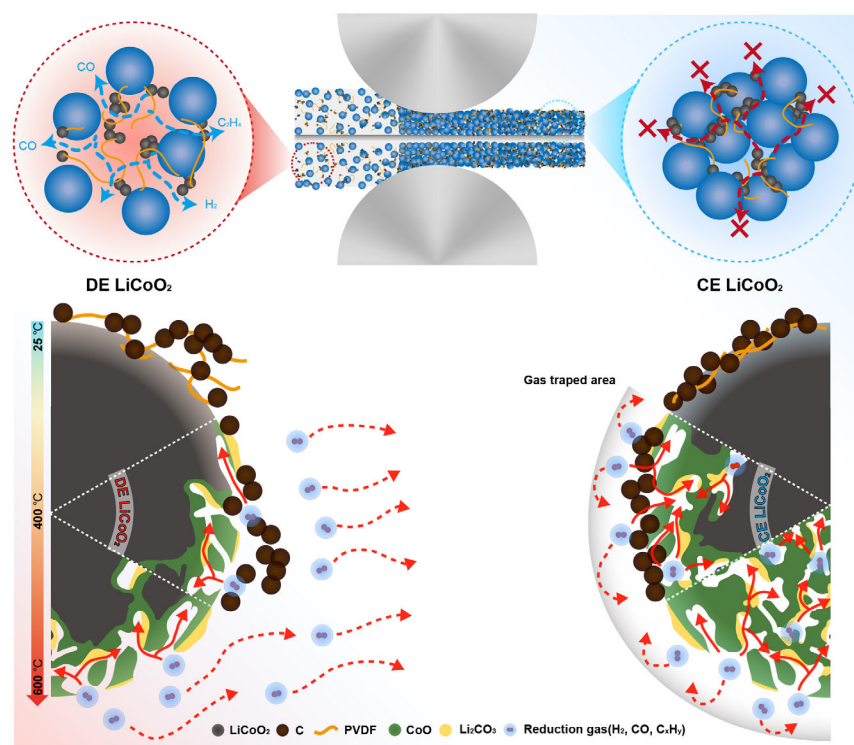


Figure 1. Schematic carbothermic reduction mechanism of DE and CE LiCoO₂. CE, calendered electrode; DE, dried electrode.

reactions.^[38–40] Solid–solid reactions can be enhanced by increasing the number of particle contact points, while solid–gas reactions can be facilitated by raising the concentration of the reducing gases. Key factors influencing these reactions include component composition and composite density. The spent cathode electrodes inherently contain conductive carbon and a Polyvinylidene fluoride binder, both capable of reducing the spent cathode materials.^[41] Their high-density composite form minimizes porosity, maximizing contact between reductants and enabling efficient reduction with less energy.^[42–44] Following the SDCR process, CoO and Li₂CO₃ are reduced from spent cathode materials, achieving over 98% recovery rates for Co and Li. These reduced materials were used as precursors to synthesize LiCoO₂ and single-crystal Ni-rich cathodes. The recycled and upcycled cathodes demonstrated electrochemical performance equivalent to that of commercial materials. This strategy simplifies the recycling process by simultaneously peeling and reducing spent cathode materials while also enabling the synthesis of upcycled single-crystal Ni-rich cathodes through a simple solid-state reaction. Consequently, this method offers a commercially viable, closed-loop recycling and upcycling solution for spent cathode materials (Figure 1).

2. Results and Discussion

2.1. Reduction Behavior of Spent Cathode Materials under SDCR Conditions

Figure 2a presents a Scanning Electron Microscope (SEM) image of ion-milled cross-sections of a calendered spent cathode electrode.

Energy-Dispersive Spectroscopy C 1s analysis confirms the uniform distribution of carbon-based compounds, such as conductive material and binder, around the spent cathode material. These compounds could serve as potential reducing agents. Figure 2b and Figure S1a, Supporting Information, show SEM images of the spent cathode material before and after washed treatment, respectively. In Figure 2b, the active material particles are uniformly mixed with conductive additives and binder, whereas in Figure S1a, Supporting Information, these surface components have been removed through washed treatment, allowing clearer observation of the active material morphology. Figure 2c displays the SEM images of the spent cathode material after the SDCR process. Figure 2c, which represents the sample immediately after the SDCR process, shows small crystalline particles dispersed on the surface. In contrast, the washed sample in Figure S1b, Supporting Information, exhibits the removal of these small particles and reveals a porous structure, distinct from that observed in Figure S1a, Supporting Information.

The recovery rates of Co and Li were calculated using the X-ray diffraction (XRD) reference intensity ratio of the SDCR-treated material and the mass of these compounds extracted through water leaching based on the equation

provided (Table S1, Supporting Information). These rates were analyzed under varying reduction temperatures and times. Figure 2d,e illustrates that the spent cathode electrode underwent heat treatment at different temperatures for 30 min. At 400 °C, only the crystalline peaks corresponding to SLCO were observed, indicating that no reduction of LiCoO₂ occurred at this temperature. When the temperature increased to 500 °C, partial reduction of LiCoO₂ to CoO and Li₂CO₃ occurred, and the recovery rates of Co and Li were 48.3% and 36.6%, respectively. At 600 °C, LiCoO₂ was completely reduced to CoO and Li₂CO₃, with traces of metallic Co detected. The recovery rates peaked at 99.5% and 98.6% for Co and Li, respectively. Figure 2f,g shows the effects of reduction time and that the conversion of spent cathode materials into different products is influenced by the retention of temperature at 600 °C. When the reduction time increases from 10 to 30 min, the recovery rates of Co and Li improved from 99% and 98.1% to 99.5% and 98.6%, respectively. However, extending the duration beyond 30 min led to a decline in the recovery rates of Co and Li, dropping below 96% and 74%, respectively. This decrease is attributed to the secondary reaction of CoO and Li₂CO₃, forming the Li₆CoO₄ phase, as confirmed by the XRD results (Figure 2f).^[45,46]

Figure S2, Supporting Information, presents the XRD data for the Co and Li compounds recovered via water leaching. The Co-based compound, comprising CoO with trace amounts of Co, was separated using a membrane filter (Figure S2a, Supporting Information). After Li⁺ ions and CO₃²⁻ remained in the leachate, lithium carbonate (Li₂CO₃) was precipitated by evaporating the water (Figure S2b, Supporting Information). The XRD data confirm that the spent LiCoO₂ material was completely reduced via the SDCR process, enabling the stable separation of Co and Li into raw material forms.

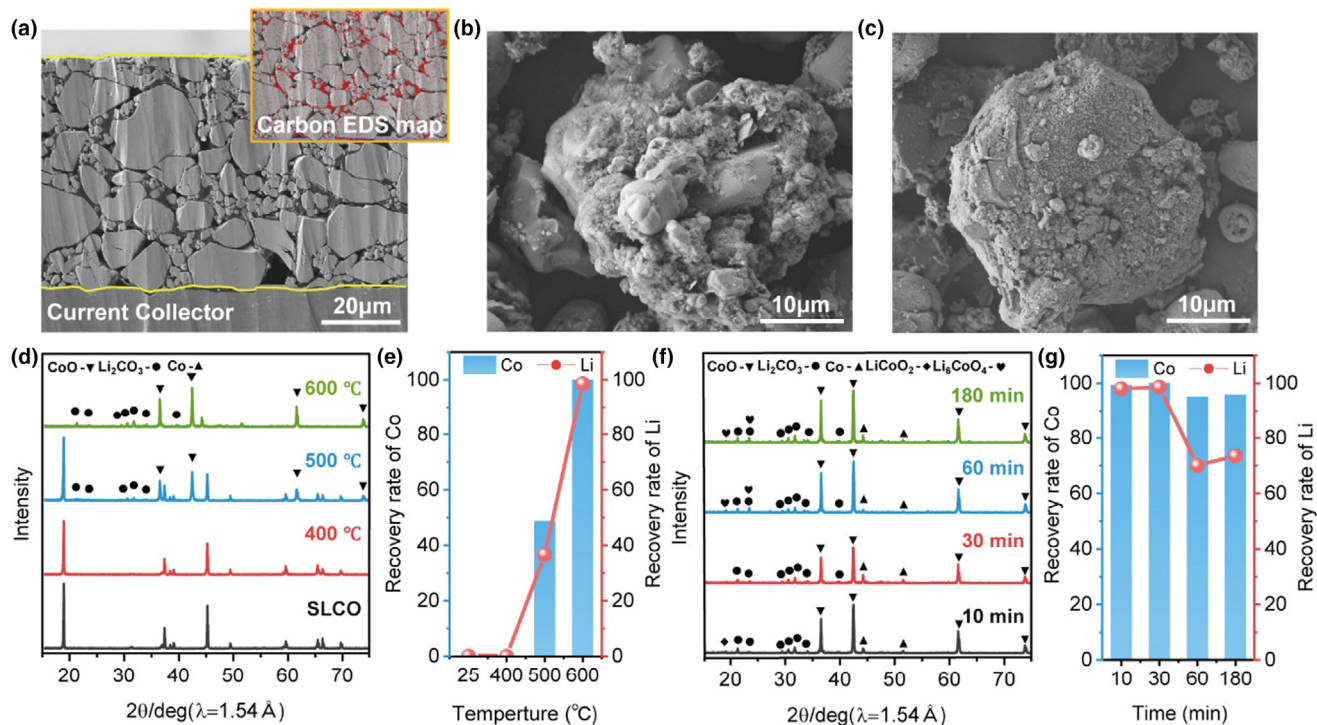


Figure 2. a) Cross-sectional SEM and EDS mapping images of a used LiCoO₂ electrode. SEM images of b) spent cathode material and c) reduced recovery cathode. XRD patterns of products obtained from the used LiCoO₂ electrode, showing the d) effect of reduction temperature (retention time = 30 min) and e) retention time (reduction temperature = 600 °C). Effect of f) reduction temperature and g) retention time on the recovery rate of Co and Li.

2.2. Revealing Thermochemical Reaction Mechanism

To investigate the effects of calendaring on the reduction behavior of high-density electrodes, two electrode types were fabricated. The first, referred to as the dried electrode (DE), was prepared by drying the

slurry on aluminum foil. The second, referred to as the calendared electrode (CE), underwent calendaring after drying. **Figure 3a,b** show cross-sectional SEM images of the DE and CE samples, with red regions highlighting the pores within the electrode. The DE sample showed a total porosity and average pore size of 10% and 11 μm, respectively. In contrast, the CE exhibited enhanced particle contact, resulting in a significantly reduced porosity of 2.5% and an average pore size of 3 μm.

Figure 3c,d shows the thermogravimetry-differential scanning calorimetry (TG-DSC) results obtained below 600 °C, a temperature range where the influence of aluminum foil is negligible.^[47] The weight change observed between 30 and 400 °C is attributed to moisture removal. Beyond 400 °C, the reduction of LiCoO₂ proceeded in distinct stages, marked by two significant weight changes. In the first stage, an exothermic peak appeared in the TG-DSC curves, corresponding to a weight loss attributed to the volatilization of the organic binder. The observed weight losses were 1.31% and 1.23% for DE and CE, respectively, indicating slightly greater weight loss in DE. This difference is attributed to the higher concentration of cracked gases (CH₄ and H₂) decomposition products of the organic binder in CE.^[48] The smaller interparticle pores in CE suppressed the

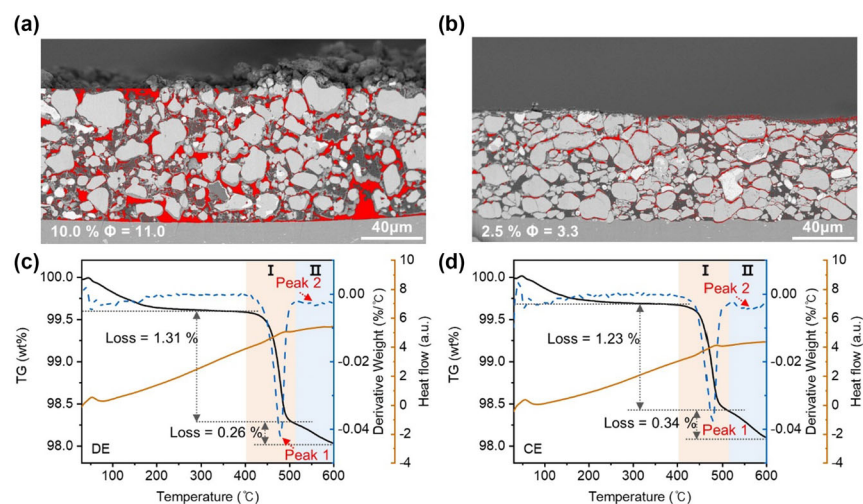


Figure 3. Cross-sectional SEM images of a) DE and b) CE LiCoO₂ electrode and porosity calculation using ImageJ analysis software. TG – DSC curve of the pyrolysis reduction of c) DE and d) CE LiCoO₂ electrodes.

decomposition reaction compared to that in DE. This suppression can be explained by Le Chatelier's principle: an increase in the concentration of products disrupts the dynamic equilibrium, prompting a shift in the equilibrium position toward the reactants to counteract the change.^[49,50] In the second stage, the weight change and absorption peak (peak 2) indicate a reaction between reducing agents (cracked gases and conductive carbon) and LiCoO_2 , as well as the continued decomposition of the organic binder.^[51,52] This process involves solid–solid and solid–gas reactions. In the solid–solid reaction, LiCoO_2 is reduced to Li_2O and CoO , releasing CO and CO_2 gases. In the solid–gas reaction, LiCoO_2 is reduced by gases (such as CO , CH_4 , and H_2) generated from the decomposition of the organic binder and conductive carbon.^[53] Under CE conditions, where smaller interparticle pores are present, these gases can accumulate at higher concentrations or undergo Knudsen diffusion, leading to more frequent interactions with surrounding particles and increased rate of solid–gas reactions.^[54–57] Consequently, CE exhibited a slightly larger weight loss (0.34%) than that of DE (0.26%). This difference is associated with an increased gas generation owing to more frequent reduction reactions in CE, as well as the volatilization of residual organic binder.

To analyze the gas evolution behavior during thermal treatment, thermogravimetry–gas chromatography–mass spectrometry (TG–GC–MS) analysis was conducted using black powder (BP) and pelleted black powder (PBP), corresponding to the dried electrode (DE) and calendared electrode (CE), respectively. To eliminate interference from the Al current collector during gas analysis, samples were prepared without the Al foil. To ensure that the thermal behavior of these samples reflects that of the DE and CE, TG–DSC analysis was also performed. As shown in Figure S3, Supporting Information, weight changes were monitored up to 700 °C. The weight loss profile below 600 °C closely matched the thermal behavior shown in Figure 3c,d. Above 600 °C, the BP sample exhibited a more pronounced weight loss. As shown in Figure S4, Supporting Information, TG–GC–MS analysis revealed that H_2O was primarily generated through reactions involving reductive gases such as H_2 and C_xH_y , which were formed from PVDF decomposition. These reductive gases were observed to evolve at higher temperatures in the PBP sample than in the BP sample, leading to a delayed H_2O evolution peak in the PBP. This suggests a shift in the decomposition kinetics caused by differences in porosity between BP and PBP. Although CO_2 evolution profiles were generally similar across both samples, a notable deviation appeared above 550 °C. In this temperature range, the PBP sample showed enhanced reactivity, facilitating the conversion of CO_2 to Li_2CO_3 via reaction with Li_2O , rather than CO_2 being released as a gas phase product.^[58] This reaction is considered a key factor contributing to the relatively smaller mass loss in the PBP sample above 600 °C.

Figure S5, Supporting Information, illustrates the XRD pattern data, showing that the CE and DE underwent identical reduction heat treatments at 600 °C. These are denoted as reduced calendared electrodes (RCE) and reduced dried electrodes (RDE). Unlike RCE, the RDE results showed that a substantial amount of LiCoO_2

remained unreduced. To investigate the residual effects of reducing gases, CE was subjected to reduction heat treatment in a vacuum atmosphere, where reducing gases were less likely to remain and could diffuse more easily. This condition is denoted as a reduced calendared electrode in a vacuum (RCEV). The results showed that LiCoO_2 remained largely unreduced. Additionally, the cross-sectional SEM image of CoO recovered during the SDCR process (Figure S6, Supporting Information) revealed a transformation into a porous structure. Table S1, Supporting Information, presents the BET data for spent LiCoO_2 (SLCO) and reduced CoO (RCoO). RCoO exhibited a significantly higher surface area (increased from 0.0762 to 6.0143 $\text{m}^2 \text{g}^{-1}$) and pore volume (increased from 0.000334 to 0.014527 $\text{cm}^3 \text{g}^{-1}$) than that of SLCO. These findings suggest that the CE environment facilitates the effective reduction of LiCoO_2 in the bulk region during the SDCR process, enabling efficient electrode recycling without the need for external reducing agents.

2.3. Electrochemical Performance of Re-Upcycled Cathodes

For RCoO, which contains Co in mixed oxidation states, an oxidation heat treatment was applied to convert it into Co_3O_4 . The resulting large porous particles were then ground via mechanical milling. Figure S7, Supporting Information, shows the XRD data for oxidized Co_3O_4 , while Figure S8, Supporting Information, displays SEM images of oxidized and mechanically milled Co_3O_4 . Figure S9, Supporting Information, provides an SEM image of the recovered LiCoO_2 produced using recycled Co_3O_4 and Li_2CO_3 . Figure 4a illustrates the XRD pattern of the recovered LiCoO_2 (RLCO), resynthesized from recycled Co_3O_4 and Li_2CO_3 . The RLCO exhibits a layered $\alpha\text{-NaFeO}_2$ structure, aligning with the standard LiCoO_2 under the space group R-3m.^[59,60]

The charging and discharging curves of spent LiCoO_2 (SLCO) and RLCO are compared (Figure 4b). At 4.3 V, the SLCO and RLCO

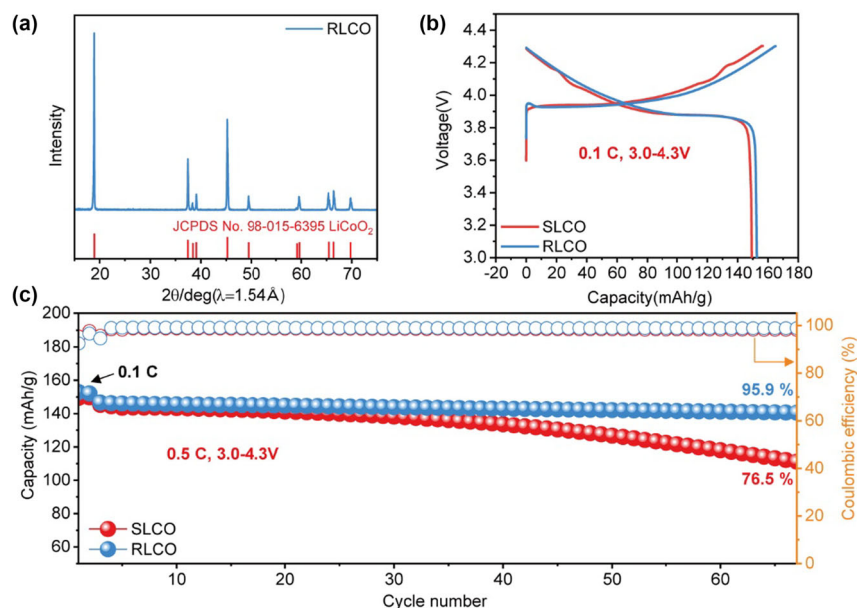


Figure 4. a) XRD patterns of the products obtained from RLCO b) Charging and discharging curves of SLCO and RLCO at 4.3 V c) Cycling performances of SLCO and RLCO at 4.3 V.

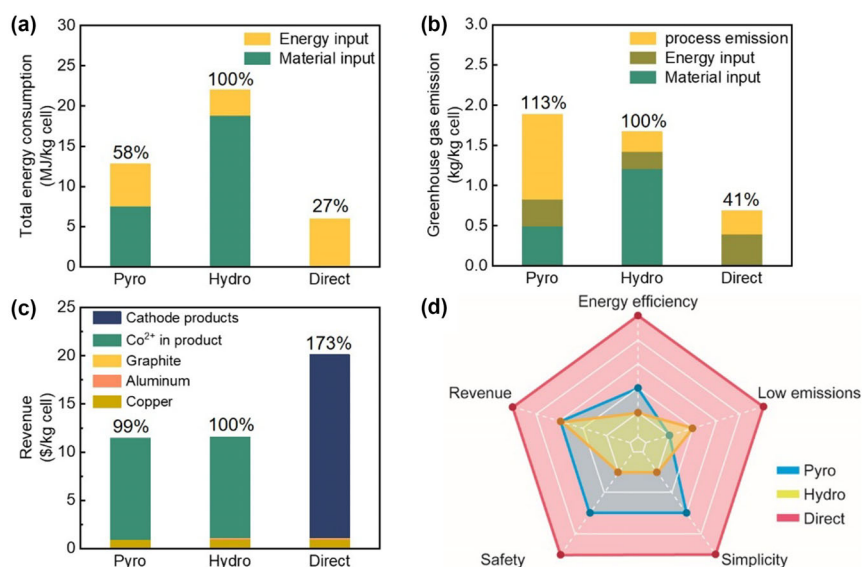


Figure 5. Economic and environmental analysis of SDCR (Direct) and other recycling approaches. a) Energy consumption, b) GHG emissions, and c) revenue for SDCR, pyro, and hydro. d) Comprehensive comparison of different recycling approaches.

delivered initial capacities of 149.4 and 152.7 mAh g⁻¹, respectively, with RLCO showing a higher initial capacity. Figure 4c presents the cycle performance of SLCO and RLCO, where SLCO exhibited a capacity retention rate of 76.5%, while RLCO achieved a significantly higher retention rate of 95.9%. These findings indicate that the resynthesis process enables the stable recovery of the layered crystal structure in RLCO, resulting in improved electrochemical performance compared to that in SLCO.

The upcycled single-crystal NCM 811 cathode material (USNCM) was synthesized using the same method reported previously. SEM images in Figure S10, Supporting Information, confirm its successful synthesis in a single-crystal form. Figure S11a, Supporting Information, displays the XRD pattern of the USNCM, which was resynthesized from recovered Co₃O₄ and a commercial Ni-Mn-Li compound. The USNCM exhibited a layered α -NaFeO₂ structure, consistent with the standard NCM under the space group R-3m. Figure S11b, Supporting Information, illustrates the charging and discharging curves of commercial polycrystalline NCM 811 (CNCM) and USNCM. The CNCM and USNCM delivered initial capacities of 194.3 and 192.1 mAh g⁻¹ at 4.3 V, respectively. Figure S11c, Supporting Information, presents the cycle performance, where the CNCM and USNCM showed a capacity retention rate of 80.7% and 94.0%.

2.4. Economic and Environmental Analysis

The economic and environmental effects of the SDCR (direct) process were evaluated using the EverBatt model and compared with conventional pyrometallurgical (pyro) and hydrometallurgical (hydro) recycling processes for LIBs. Key factors such as recycling efficiency, carbon neutrality, and economic viability including energy consumption, greenhouse gas (GHG) emissions, and revenue were calculated under conditions specific to LiCoO₂ battery recycling, with the widely adopted hydrometallurgical process serving as a baseline. The

Supporting Information (Figure 5 and Figures S12–S14; Tables S3 and S4) provides details of the evaluation process using the EverBatt model.^[61] Figure 5a shows a comparison of the total energy consumption of each process. The hydro process, which involves complex leaching and precipitation steps, requires various raw materials, resulting in high material input and an energy consumption of 22.02 MJ kg⁻¹. The pyro process, with its relatively lower material input, exhibited an energy consumption of 12.8 MJ kg⁻¹. In contrast, the SDCR process demonstrated significantly lower energy consumption at 6 MJ kg⁻¹, equivalent to only 27% of the energy required for the hydro process. This reduction is attributed to the streamlined approach of the SDCR process, which relies on heat treatment without additional material inputs. Figure 5b presents a comparison of the GHG emissions associated with the recycling processes. The hydro and pyro methods exhibited high GHG emissions owing to their substantial energy and material requirements. The pyro process produced significantly higher GHG emissions during smelting, exceeding those of the hydro process by 113%. In contrast, the direct process required no additional material input, resulting in emissions comparable to those of the hydro process but achieving a 41% reduction in GHG emissions overall. Figure 5c illustrates the revenue generated by each recycling method, with the direct process showing the highest revenue owing to its direct upcycle of used cathode materials into new, high-value cathodes. Figure 5d shows key metrics, including energy efficiency, GHG emissions, profitability, safety, and process simplicity. Regarding safety, the direct process offers a lower risk than that of the hydro and pyro processes, as it avoids the use of temperatures above 1000 °C and eliminates hazardous chemicals. Moreover, the SDCR process is simple and more straightforward.

3. Conclusion

The SDCR process proposed in this study provides a promising approach for recycling cathode materials by eliminating the need for external reducing agents. By leveraging the carbon conductive materials and organic binders present in the electrode, the method reduces costs and simplifies the recycling process by omitting the addition of reducing agents and associated mixing steps. Furthermore, the calendaring procedure enhances particle contact and minimizes pore space, thereby promoting the reduction reaction. The TG-DSC analysis revealed that CE showed a greater weight change and enhanced reduction during heat treatment compared to that of DE. This phenomenon is due to the formation of closed pores between particles in CE, which effectively trap reducing gases. To confirm these findings, XRD analysis was conducted on heat-treated samples of RDE, RCE, and RCEV under identical temperature conditions. The results showed that in RDE, approximately half of the LiCoO₂ remained unreduced, while RCE exhibited enhanced reduction due to the presence of residual reducing gases. However, in RCEV, despite improved interparticle contact and heat treatment under conditions minimizing internal gas residues, more LiCoO₂ remained

unreduced compared to that of RDE. These findings suggest that a high-density electrode environment retains reducing gases, thus facilitating the reduction reaction. The SDCR process, which avoids the addition of reducing agents, offers potential solutions to address key challenges in recycling LIBs, including environmental effects, cost reduction, and process efficiency. Since the presence of confined pores promotes the reduction reaction, this approach is expected to be applicable to various types of spent cathode electrodes. However, its effectiveness is constrained by insufficient consideration of various variables that may arise during the practical recycling process.

4. Experimental Section

Simplified direct carbothermic reduction: The cathode electrode scraps used in this study consisted of active material, conductive agent, and binder in a weight ratio of 94:3:3. The electrodes were double-sidedly coated, with a loading of approximately 47 mg cm^{-2} and a composite density of about 3.7 g cm^{-2} . The cathode electrode scraps were detached from the Al foil using an ultrasonicator with NMP, followed by separation of the spent cathode materials with a centrifugal separator. The materials were thoroughly rinsed several times with dimethyl carbonate and ethanol to remove impurities. Finally, the cleaned materials were freeze-dried to obtain the final product. For investigating the effect of electrode composite density, CE (calendered electrodes) and DE (dried electrodes) were prepared. For controlled variable analysis, the cathode material was separated from the cathode scrap, and new electrodes were fabricated under the same conditions as the cathode electrode scrap. The cathode electrode scraps were heat-treated under an argon atmosphere with a heating rate of 5°C min^{-1} . The temperature variables were 400, 500, and 600°C , with a 30 min treatment at each temperature. For time variables, heat treatment was conducted at 600°C for 10, 60, and 180 min. To assess the impact of residual gases, the RDE (reduce the dried electrode) and RCE (reduce the calendered electrode) were heat-treated under an argon atmosphere at 600°C for 10 min with a heating rate of 5°C min^{-1} . In the case of RCEV (reduce the calendered electrode in a vacuum), the treatment was conducted under the same conditions but in a vacuum atmosphere. The SDCR products were subjected to water leaching and separated by filtration. For the leaching process, 250 mL of distilled water was used per 5 g of SDCR products, with stirring at 400 RPM using a magnetic bar for 1 h. The Co-based compound was filtered using a membrane filter, and lithium carbonate was obtained by evaporating the water from the leachate.

Synthesis of re-upcycling cathodes: The recovered Co-based compound was heat-treated at 700°C for 1 h to oxidize it to Co_3O_4 , followed by mechanical milling to break it into smaller particles. For the recovered LiCoO_2 , the recycled Co_3O_4 and Li_2CO_3 were mixed using a mixer to achieve a Li/Co molar ratio of 1.03. The mixed powder was then subjected to a two-step heat treatment in a box furnace. Initially, it was heated at a rate of 5°C min^{-1} under an air atmosphere at 600°C for 5 h, followed by a temperature increase to 950°C , where it was heat-treated for 12 h to complete the synthesis. Commercial NCM cathode materials were purchased from Alibaba. The upcycled NCM single-crystal cathode material was fabricated using the recycled Co_3O_4 , following the same synthesis method as previously developed by our research group for Ni-rich single-crystal cathode materials.^[62]

Material characterization: To investigate the removal of conductive materials and binders within the cathode composite as a function of temperature, thermal analysis was conducted using a Thermal Analyzer (TA Instruments/Discovery DSC 25, Discovery TGA 55, TMA Q400) at a heating rate of $10^\circ\text{C min}^{-1}$. To investigate the gas evolution behavior associated with the decomposition of conductive agents and binders in the cathode composite, thermogravimetry-gas chromatography–mass spectrometry (TG, STA 449 F5 Jupiter, NETZSCH; 5977B GC/MSD, Agilent) was performed using helium (He) as the carrier gas at a heating rate of $10^\circ\text{C min}^{-1}$. TG–GC–MS analysis was carried out accordingly. The morphology of the particles and electrode cross-sections was observed using a Scanning Electron Microscope (SEM, S-2400, Hitachi). To analyze the distribution of conductive materials and binders within the electrode, Energy-Dispersive X-ray Spectroscopy (EDS, Octane elect EDS system, AMETEL EDAX) was employed. For analyzing cross-sectional electrode images, Cross-section &

Planar milling system (697 lion II, Gatan) was conducted to obtain the cross-sectional electrode. A Brunauer Emmett teller (BET, HM model-1208, Macsorb) analysis was conducted to evaluate the specific surface area and pore formation of the reduced materials. The crystalline structure of the spent cathode materials, SDCR products in their reduced state, and re-upcycled cathode materials was analyzed using X-ray Diffraction (XRD, D/Max-2200/PC, Rigaku). The total porosity and average pore size in the electrode cross-section SEM images were calculated using ImageJ analysis software.

Electrochemical characterization: The cathode slurry for electrochemical evaluation was prepared with a weight ratio of 94:3:3 for cathode active material (LiCoO_2 or $\text{LiNi}_{0.8}\text{Co}_{0.1}\text{Mn}_{0.1}\text{O}_2$), conductive agent (Carbon black, Super-P), and binder (PVDF, Solef 6020). The slurry was loaded onto an aluminum foil at $11 \pm 0.5 \text{ mg cm}^{-2}$ and calendered using a roll press to achieve a composite density of 3.6 g cm^{-3} . The fabricated electrodes were dried in an oven at 120°C for 6 h. Cell fabrication was carried out in an inert atmosphere glove box using CR2032 coin cells. For the anode, a 1 mm thick Li foil was used, and the electrolyte consisted of 1.3 M LiPF_6 dissolved in a mixture of EC, EMC, and DMC (3:4:3 in volume). Electrochemical performance was evaluated using galvanostatic charge–discharge testing within a voltage range of 3.0–4.3 V (vs Li/Li^+). For initial charge–discharge cycles, a constant current of 0.1 C was applied for charging to 4.3 V, followed by a constant voltage charge until the current reached 0.05 C cut-off. Discharge was performed at 0.1 C to 3.0 V. For cycling tests, charging was done at 0.5 C under constant current, followed by constant voltage charging to a 0.05 C cut-off. Discharge was carried out at 1 C.

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Conflict of Interest

The authors declare no conflict of interest.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

Keywords

carbothermic reduction, closed-loop recycling, lithium-ion batteries, simplified direct carbothermic reduction, spent cathode material

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