

Sustainable Pre-treatment Method for Lithium-ion Production Scrap Material Recovery and Re-utilisation

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Demand for Lithium-ion batteries (LIB) is expected to grow from 600 GWh in 2022 to 4000 GWh by 2030 leading to the opening of numerous new battery gigafactories worldwide (IEA, 2024). The production of batteries includes steps where a notable portion of electrode production will be discarded as scrap due to processes like electrode cutting or quality controls. This scrap production rate in battery production can range from 5% to 30% depending on the maturity of the production process (Orangi et al., 2024). Especially the factories in the start-up phase typically struggle with high scrap rate. Therefore the heavy increase in the number of production lines starting their operation has made scrap materials the main supply to battery recycling sites surpassing the volume of End of Life (EoL) Batteries.

Battery production scraps fundamentally consist of the same materials as EoL batteries that are considered conventional material flow in recycling. However, the properties of these materials are significantly different. The key difference is that scrap electrodes have not been assembled in batteries and hence the material is not degraded by exposure to electrolyte nor cycling and most of the safety risks related to battery pack recycling are avoided. Conventional battery recycling is designed to deal with the characteristics and hazards related to recycling of EoL battery packs and therefore it is based on destructive methods where high temperatures and aggressive leaching are used to recover most valuable elements. However, destructive approach is not necessary for the recycling of intact scrap material.

This study focuses on the direct recycling of cathode production scraps, a novel non-destructive recycling approach. Direct recycling includes separation and reparation of the active material and direct reuse of the material in the production of new cells. In the case of production scraps this process can be simplified as no extensive material reparation is needed. The upscaling of this kind of direct recycling of production scrap requires fast, cheap, and reliable material liberation methods that prevail the material structure and electrochemical performance. Currently solvent methods and high temperature thermal treatment are widely used in industry to decompose the adhesion created by the binder between active material and the current collector. However, these methods struggle with environmental hazards, high energy intensity and demand for complicated further processing (Shin et al., 2023). To avoid these challenges, low temperature treatments are studied in different environments.

This study investigates direct recycling of LIBs production scraps, focusing on the most common cathode chemistries: Lithium Nickel Cobalt Manganese oxides (NMC) and Lithium Iron Phosphate (LFP) cathodes. Direct recycling was performed under various thermal treatment conditions (process temperatures: 20°C, 200°C, 300°C, 400°C, and 600°C, in air and inert N₂ atmospheres, for 30 min) followed by mechanical detachment of the cathodic material using ball milling. The recycled materials were then used to produce new LIBs coin cells to test electrochemical behavior. Energy consumption was directly measured during experiments to evaluate economic and environmental costs of the different conditions.

Active material separation was successfully achieved at 600°C in nitrogen for LFP and at 600°C in air for NMC. These temperatures are currently considered standard in the industry for active material liberation (Yan et al., 2023). The material separation efficiency using direct recycling is comparable to that of conventional recycling

methods, even at 400°C in air for both LFP and NMC, and at 400°C in N₂ for LFP (Figure 1). LFP active material separation was also possible at lower temperatures, whereas NMC cathodes required temperatures exceeding 300°C. In fact, treatment of NMC at 200°C and 300°C reduced recovery efficiency, suggesting that melting the undegraded PVDF binder can increase adhesion between the current collector and the active material.

Economic and environmental performance evaluations confirmed that direct recycling offered both economic benefits and CO₂ emission reductions compared to production from pristine active material across all conditions studied (Figure 1). However, results demonstrate how lowering the temperature from the standard 600°C to 200°C for LFP resulted in the greatest savings and CO₂ emission reductions, even with a lower active material recovery rate. For NMC, direct recycling at 400°C in air proved to be the most cost-effective and environmentally friendly option. New cells were successfully produced from the material treated at 400°C, as this condition performed well for both LFP and NMC. LFP material treated in 400°C in nitrogen achieved a discharge capacity of 150 mAh/g and a coulombic efficiency above 98%. In contrast, NMC direct recycling still requires further optimization due to structural changes that occur during thermal treatment.

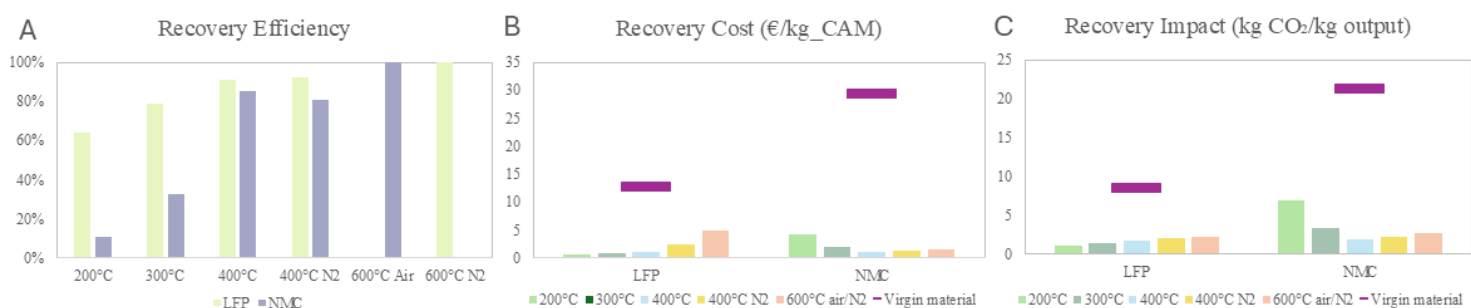


Figure 1 A) Recovery efficiency in different conditions. B) Cost (€/kg_CAM) of direct recycling in different temperatures and active material production cost from virgin materials. C) Recovery Impact (kg CO₂/kg output) of direct recycling in different temperatures and active material production impact from virgin materials.

In conclusion, this study demonstrates that low-temperature thermal treatment can enhance the economic and environmental sustainability of recycling production scraps while maintaining electrochemical performance, especially for LFP material. This approach offers a promising avenue for future research and industrial application in lithium-ion battery material recycle.

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