Study of Electrode Materials for Li-ion Batteries Obtained through Direct Recycling

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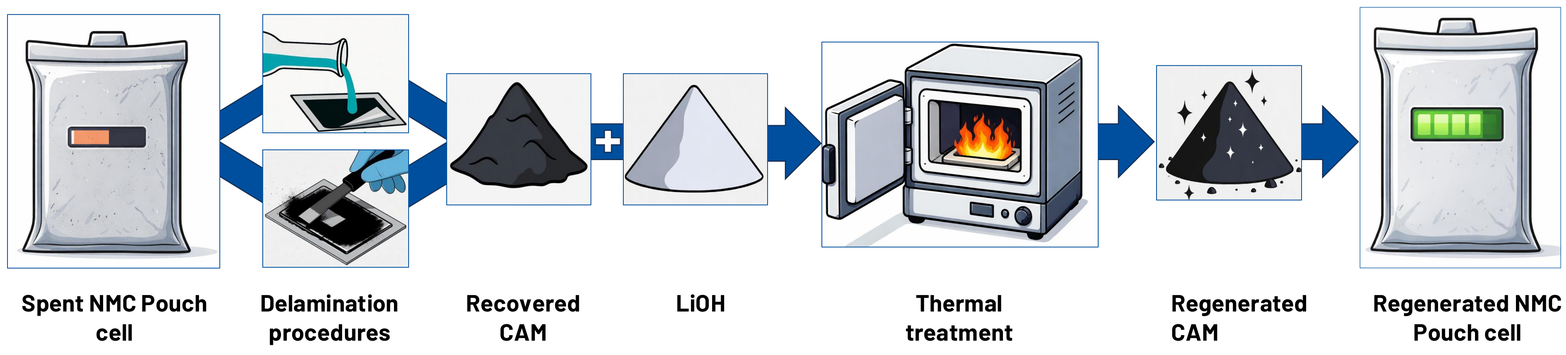
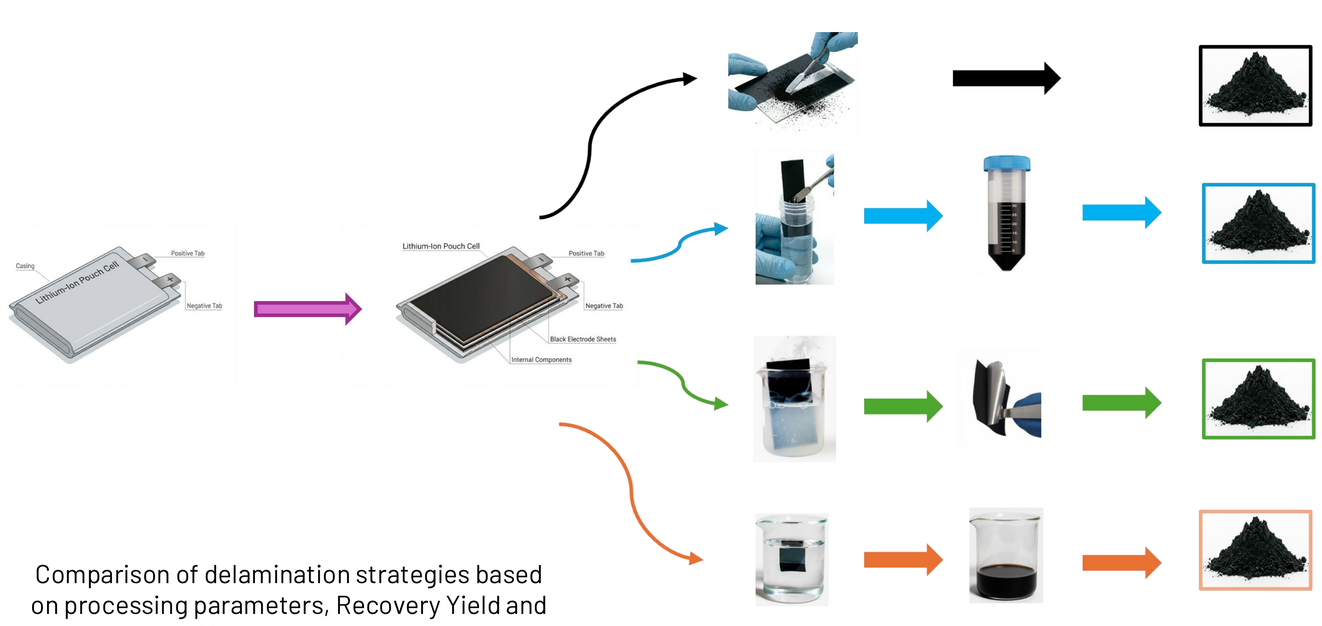
Introduction

Layered transition-metal oxide cathodes, such as $LiNi_xMn_yCo_zO_2$ (NMC), rely on critical elements, that are geographically concentrated, energy-intensive to extract, and subject to geopolitical risk. Many existing recycling approaches focus primarily on the recovery of secondary commodities enriched by a single metal. By preserving the original cathode structure and composition, Direct recycling seeks to retain the embodied energy, crystallographic order, and morphological features of spent electrodes. Processes such as delamination, re-lithiation, enable the regeneration of cathode materials with performance approaching that of pristine materials, while substantially reducing environmental impact.



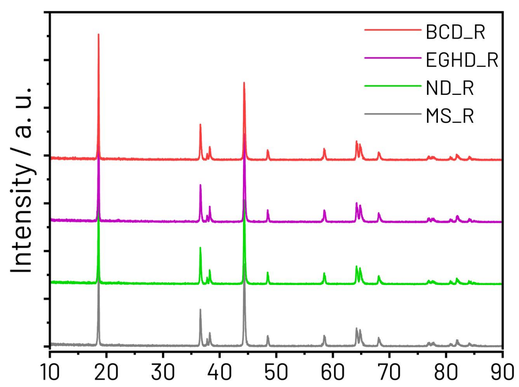
Delamination Procedures



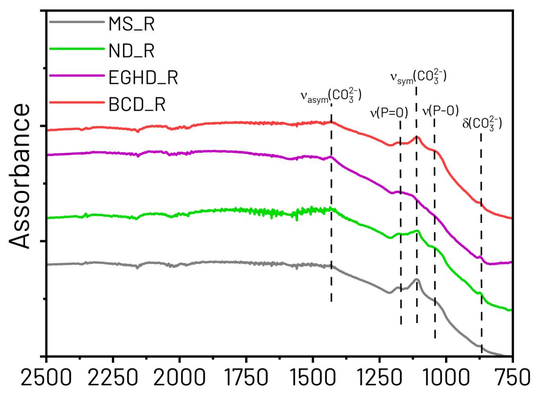
Comparison of delamination strategies based on processing parameters, Recovery Yield and Al contamination

Sample	Main Mechanism	Recovery (%)	Al (wt.%)
MS	Abrasive detachment	73	0.93
ND	Chain disentanglement	80	1
EGHD	H-bond disruption	89	0.5
BCD	Plasticisation + H-bond disruption	89	0.38

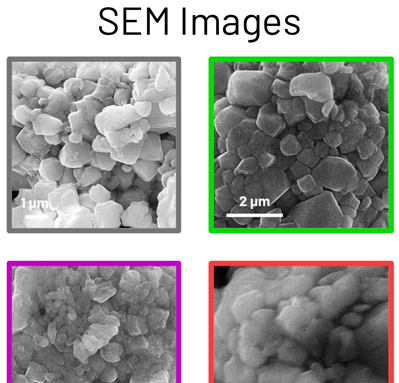
XRD



FTIR-ATR

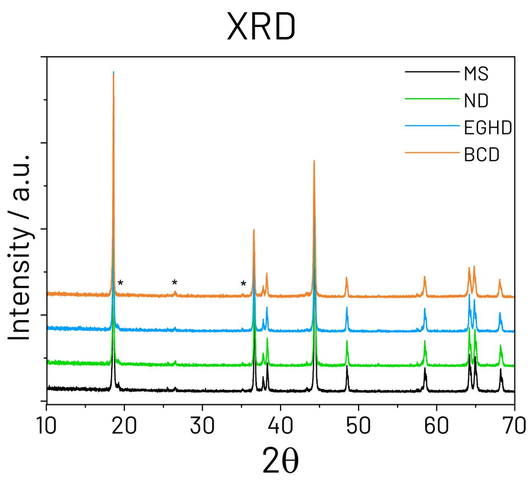


SEM Images

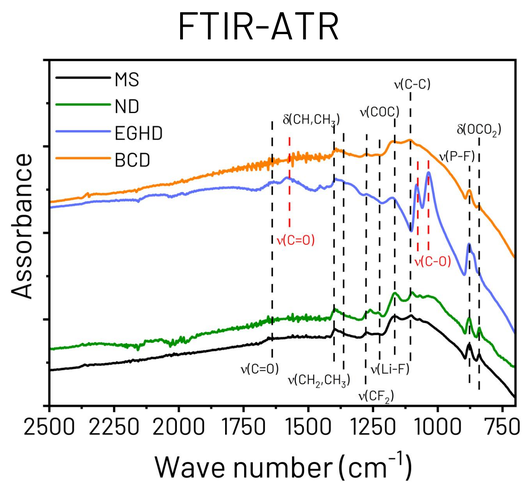


- The XRD confirms full restoration of the rhombohedral layered framework, consistent with a stable hexagonal lattice.
- FTIR spectra reveals the persistence of residual surface compounds after regeneration.
- Morphological analysis reveals homogeneous morphologies across all regenerated samples, consisting of polyhedral primary crystallites assembled into micrometre-sized secondary particles.

XRD



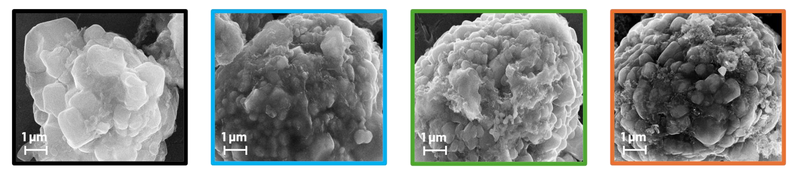
FTIR-ATR



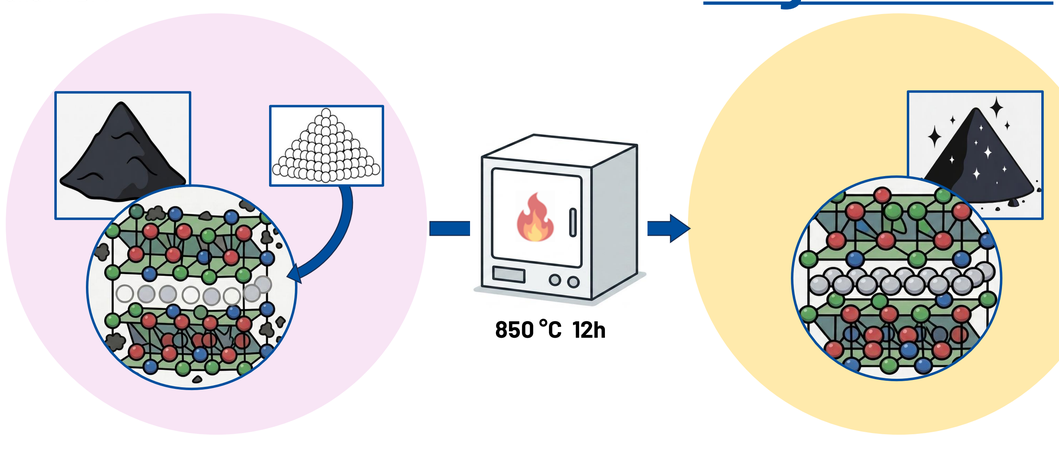
- The XRD patterns collected from all samples can be indexed to a layered α-$NaFeO_2$-type structure with rhombohedral symmetry (space group R-3m), in good agreement with literature reports for NMC442 layered cathodes. A few weak additional peaks are observed that can be attributed to residual graphite and alumina.
- FTIR-ATR spectroscopy allows to investigate the nature of organic species associated with the recovered cathode materials. All samples exhibit spectral features associated with residual electrolyte, binder, and interphase species.

SEM Images

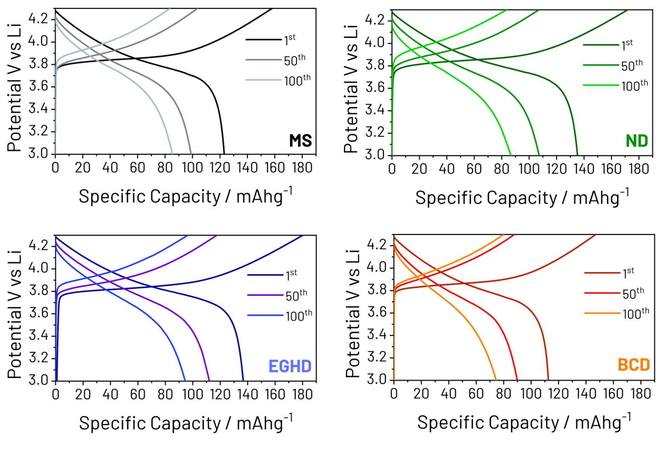
All powders exhibit the characteristic polyhedral NMC aggregates (100-300 nm sub-units), while locally coated regions indicate the residual Super P and PVDF.



Regeneration



Galvanostatic Cycling
C/10 (1C=130mA/g), LP30 electrolyte



The regenerated cathodes recover substantial electrochemical activity, highlighting the effectiveness of the solid-state treatment, while suggesting that additional purification or surface treatments could further improve the efficiency and long-term cycling performance.

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