Direct Recycling of End-of-Life Lithium-Ion Batteries by Electrochemical Route

J. Castro[1,2], M. Gómez[1], M.R. Bermejo[1], P. Moretti[1], M. Merlo[3], Y. Belce[3] and J.J. Biendicho[3]

[1]CARTIF Technology Centre, Area of Circular Economy, Boecillo, Valladolid, 47151, Spain
[2]Institute of Sustainable Processes, University of Valladolid, Valladolid, 47011, Spain
[3]Catalonia Institute for Energy Research (IREC), Sant Adrià de Besòs, Barcelona, 08930, Spain
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Presenting author email: juacas@cartif.es

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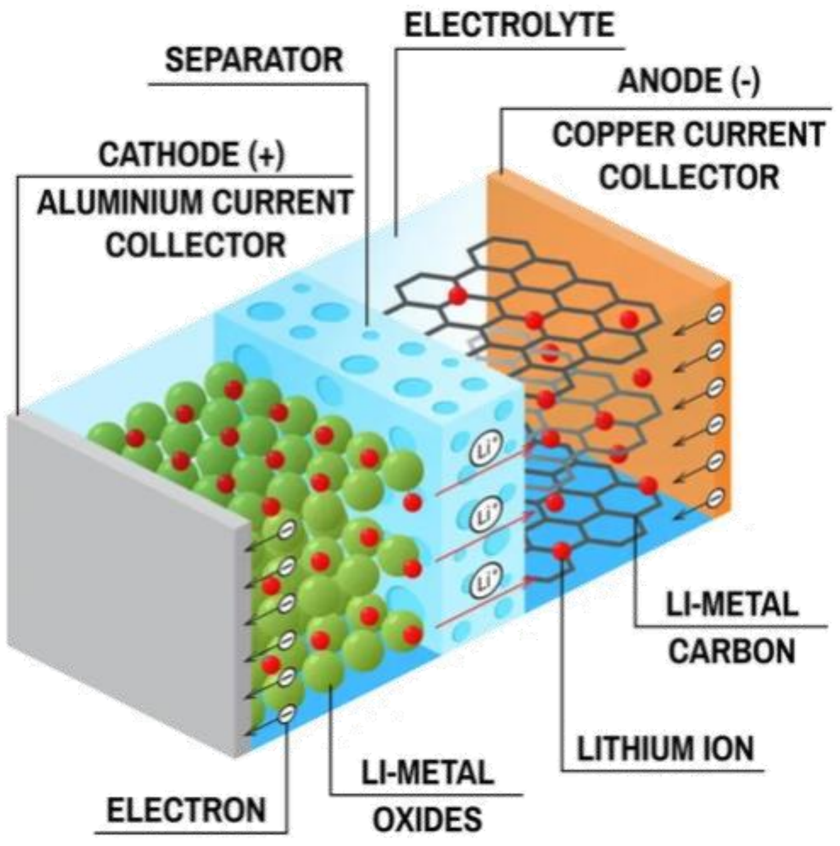
INTRODUCTION

In the search for new renewable energy solutions, **Lithium-Ion Batteries** (LIBs) are one of the most promising solutions in recent decades, offering **high energy density, long life and multidisciplinary applications**. Thanks to these advantages, LIBs have been perfectly integrated **into energy storage**, both for stationary use in homes and industry, and **for powering the electric motors of heavy vehicles or light personal transport**. The scientific community's intense research into the vagaries of lithium-ion chemistry has catalysed remarkable innovation and presented numerous paradigms to elucidate the fundamentals of LIB technologies in their ever-evolving permutations.



In LIBs, Li⁺ move effectively and reversibly between the anode and cathode through an electrolyte, assisted by separators. This process occurs during both charging & discharging. Electrode materials and electrolyte formulas advances are enhancing efficiency and sustainability of new LIBs generations.

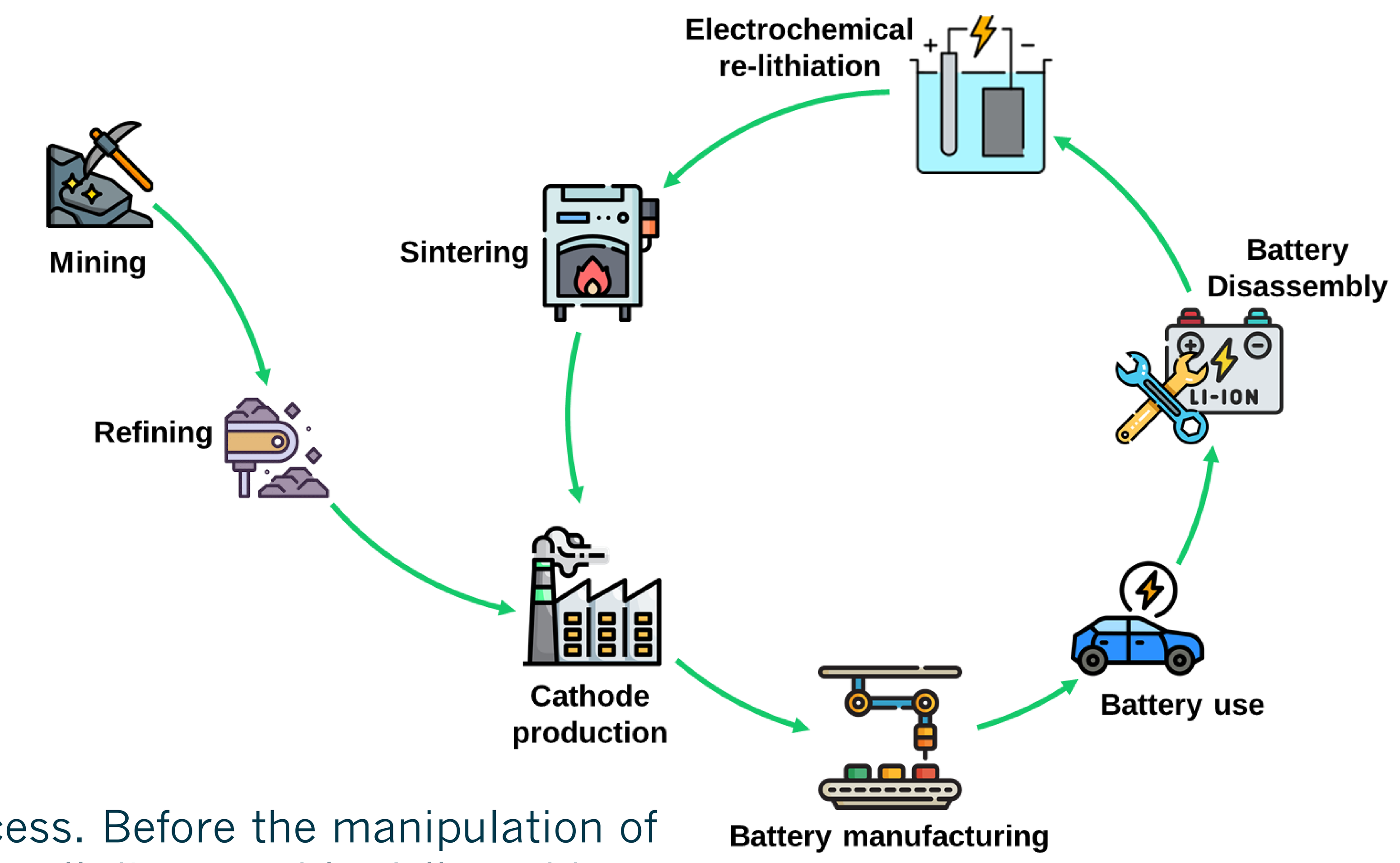
Researchers face a myriad of issues ranging from capacity degradation to safety and the environmental footprint of raw materials. In response, their **efforts are increasingly** focused on not only addressing these concerns, but also integrating **sustainability across the entire battery lifecycle**. From **advanced recycling technologies** that recover valuable materials from spent batteries to the search for **manufacturing eco-friendly alternatives**, these solutions underscore the scientific community's commitment to creating a circular LIBs value chain, where advances in design, materials and recycling technology converge **to deliver minimal environmental impact and maximum resource efficiency**.

	Pyrometallurgy	Hydrometallurgy	Direct recycling
Pros.	Effective in recovering metals like Co, Ni & Cu. **Well-established and widely used processes**	Low energy consumption. **Capable of recovering a wider range of metals, including lithium**	Preserves the original structure of battery materials. **Lower environmental impact.**
Cons.	High energy costs due to the required high temperatures. **Does not recover lithium. May generate harmful emissions**	Requires handling of hazardous chemicals. **Processes can be slow and complex**	Low TRLs. **Requires pre-sorted and well-conditioned batteries**

The battery recycling market is rapidly growing, driven by the need for critical raw materials and European policies aimed at reducing external dependencies. However, current industrial recycling facilities are insufficient for the expected volumes of end-of-life LIBs from electric vehicles over the next decade. With **global battery demand projected to increase by 25% annually to 2,600 GWh by 2030** (McKinsey & Co, 2023), enhancing recycling technologies and reducing environmental impacts are essential.

RESULTS & DISCUSSION

Direct recycling process is divided in two well-defined steps such as fine disassembly and separation, and the re-lithiation step. This study aimed to **directly recycle NMC-622 cathode electrode from the EoL LIB of an electric vehicle (EV)**, through prior efficient cell disassembly in order to achieve cathode electrodes of LGX E63B pouch cells. This NMC chemistry of LIBs currently dominates the market and is a typical layered Li transition metal oxide cathode material. Long-term use (charge-discharge) can lead to various component and structural failures such as loss of active lithium, dissolution of transition metal ions, mixing of nickel and lithium, or dendrite formation over the anode, among others, which significantly degrade the performance of the NMC cathode material (Kang, 2021; Jiang, 2021; Yin, 2021).



Fine disassembly and separation step were achieved after the deactivation process. Before the manipulation of the pouch cells, deep discharging was achieved to reduce electrical risks during cell disassembly, followed by a second safety measure of freezing to -18ºC for 24h prior to manual cell opening, which was performed in a fume hood to separate casing, anode, cathode and separator. A vacuum oven was then used to evaporate and condense the electrolyte. The re-lithiation step was carried out in a electrochemical cell formed by free-electrolyte cathode electrode which was employed as working electrode (WE), an Ag/AgCl with saturated KCl solution as reference electrode (RE) and Pt as counter electrode (CE). All three electrodes were immersed in a lithium solution. The **influence of** different **lithium solutions such as Li_2SO_4 and LiOH** at different concentrations (0.2–1M) and **variation of cathodic current density** (between -0.4 and -1 mA/cm2) **was investigated**. Once the electrochemical insertion of Li⁺ into the spent cathode electrode was completed, the cathode electrode was annealed in a muffle at 800ºC during 2h to reach the **healed cathode active material (CAM)** for the electrochemical performance. To check the viability of the recycling process, the CAM was tested in half-cell using a Celgard 2400 separator and $LiPF_6$ DEC: EC electrolyte in the voltage range 2.7 - 4.2 V vs Li metal. The figure shows how this direct recycling route fits inside the LIBs value chain.

CONCLUSIONS

The **regenerated NMC-622 cathode** material demonstrates a discharge capacity of 150 mAh/g at 0.05C during activation and 100 mAh/g at 0.1C with a capacity retention of 80% after 100 cycles. These electrochemical results **align with state-of-the-art NMC622 cathode materials that are commercially available**. The recycling approach outlined in this study is not only straightforward but also scalable, offering a potential avenue to tackle the environmental issues associated with large quantities of end-of-life lithium-ion batteries (EoL LIBs).

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