# Direct Recycling of Graphite from Spent Batteries and Production Scraps for the Development of a Circular and Sustainable Economy

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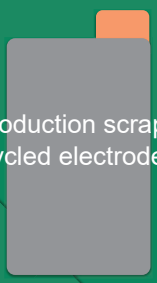
## - Context -

Current recycling routes for lithium-ion batteries (LIBs), pyrometallurgy and hydrometallurgy, do not target the recovery of graphite. This material constitutes 12-21% of the cell mass that is lost as waste. Direct recycling techniques are emerging, which target the recovery of regenerated materials ready to be reinserted in the electrode-processing loop.

## - Objective -

Optimization of a direct recycling route to recover regenerated graphite material from production scraps and cycled cells (End-of-Life, EOL, condition State-of-Health, SOH<80%) to create a sustainable circular economy and decrease material waste.

## - Results -



· Production scraps
· Cycled electrodes (EOL)

**Mild leaching** → Acid: **2 M $H_2SO_4$ + 5vol% $H_2O_2$**
Solid-to-liquid ratio: **52 g $L^{-1}$**
Leaching time: **2 h**
Leaching temperature: **70 ºC**

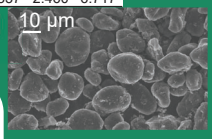
**Calcination** → Heating rate: **5 ºC $min^{-1}$**
Temperature: **400 ºC**
Holding time: **1 h**

· Regenerated graphite
· No polymeric impurities

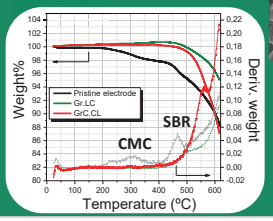
**XRD**

| | Graphitisation degree | d002 | a=b | c |
|---|---|---|---|---|
| Pristine | 86% | 0.337 | 2.463 | 6.718 |
| Scrap LC | 67% | 0.338 | 2.462 | 6.720 |
| Scrap CL | 82% | 0.337 | 2.464 | 6.717 |
| EoL CL | 84% | 0.337 | 2.460 | 6.717 |

**FE-SEM**




**TGA**




**Gr.LC**
Prod. scraps - + 
**GrC.CL**
EOL cell - + 

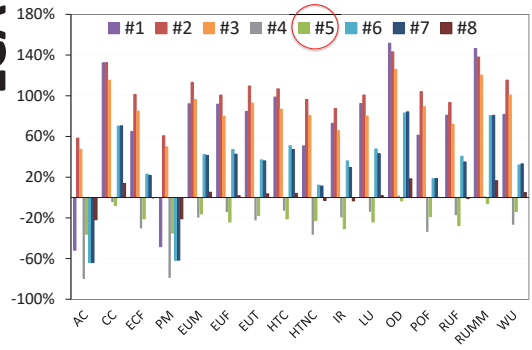
Design-of-Experiments (**DOE**) was proposed to optimise the leaching conditions.

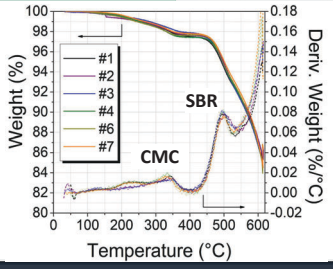
Life Cycle Assessment (**LCA**) was carried out to determine the environmental impact of each condition.

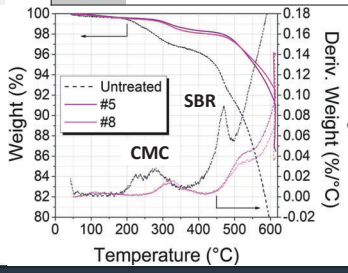
**DOE**

| #Test | S/L (g $L^{-1}$) | Temperature | Leaching time | [$H_2SO_4$] |
|---|---|---|---|---|
| #1 | 100 | 40 °C | 20 min | 0.2 M |
| #2 | 100 | 40 °C | 1 h | 1.2 M |
| #3 | 100 | 60 °C | 20 min | 1.2 M |
| #4 | 100 | 60 °C | 1 h | 0.2 M |
| **#5** | 65 | 40 °C | 20 min | 1.2 M |
| #6 | 65 | 40 °C | 1 h | 0.2 M |
| #7 | 65 | 60 °C | 20 min | 0.2 M |
| **#8** | 65 | 60 °C | 1 h | 1.2 M |

**LCA**




**Condition #5** was selected as the most sustainable and effective direct recycling protocol:

- ✓ Elimination of copper from the current collector.
- ✓ Elimination of SBR.
- ✓ Elimination of CMC.
- ✓ Lower environmental impact.

## - Conclusions -

· Direct recycling route for recycling of graphite from production scraps and EOL cells tested.
· Clean and regenerated graphite material recovered after a mild leaching and calcination.
· Leaching protocol optimised after proposed DOE and LCA study on the environmental impact of the process: **65 g $L^{-1}$**, **40 ºC** leaching for **20 minutes** with **1.2 M $H_2SO_4$**.

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