

# A Closer look on Synergetic Leaching of $\text{LiFePO}_4$ - and $\text{LiNi}_x\text{Mn}_y\text{Co}_z\text{O}_2$ - Containing Lithium-ion Battery Waste.

Johannes J.M.M. van de Ven <sup>1</sup>, Yongxiang Yang <sup>1</sup> & Shoshan T. Abrahams <sup>1\*</sup>

<sup>1</sup> Delft University of Technology, Department of Materials Science and Engineering, Mekelweg 2, 2628 CD Delft, The Netherlands.

## Abstract

The increasing need for rechargeable lithium-ion batteries (LiBs) results in a large demand for the critical raw materials (CRMs) they contain, such as Li, Mn and Co. Various combinations of elements are used in LiBs, depending on the desired application and time of manufacturing. Important examples of these are  $\text{LiFePO}_4$  (LFP) and  $\text{LiNi}_x\text{Mn}_y\text{Co}_z\text{O}_2$  (NMC). To efficiently dissolve the Li, Ni, Mn and Co for their recovery many leaching processes make use of high acid concentrations (2 mol/L  $\text{H}_2\text{SO}_4$ , 4 mol/L HCl), as well as reducing agents such as  $\text{H}_2\text{O}_2$ . However, the current study focusses on lowering the use of chemicals by simultaneous leaching of LFP and NMC from various black mass samples, ranging from pristine cathode active materials (CAMs) to multiple industrially pre-treated battery wastes. This is achieved by using the Fe(II) in LFP as a reducing agent to dissolve the Ni, Co and Mn. Results show that the addition of spent LFP can improve the leaching efficiency by up to 55%, leading to 95 – 100% dissolution even at low (0.63 mol/L  $\text{H}_2\text{SO}_4$ ) acid concentration. Results also show that there is a large effect of waste composition and pre-treatment, such as an advantageous role of impurities in the dissolution process. Both Cu and Al impurities present in industrially pre-treated battery waste were found to improve the leaching efficiencies through catalysis by  $\text{Fe}^{3+}/\text{Fe}^{2+}$ . In addition, we show that separation of the  $\text{FePO}_4$  after leaching can be easily facilitated by pH adjustment.