Supplementary information

Fe removal

First, our method was tested to separate Fe from Co: 0.2262g of $LiFePO_4$ and 0.2343g of $LiCoO_2$ were mixed in 20mL of 4M H_2SO_4 and 4mL of H_2O_2 at 74℃ for 2hrs. Undissolved $LiFePO_4$ was filtered from solution and NaOH was added to the solution until the pH=3.5. $Fe(OH)_3$ precipitate was separated from the solution by filtration and tested with SEM and EDS. The results (Figure 1) showed that there was no Co in the precipitate. The concentration of Fe in the solution, after filtration, was tested by atomic absorption spectroscopy (AAS). No Fe was detected in AAS measurement. Therefore, the AAS result demonstrated that Fe was successfully separated from Co.

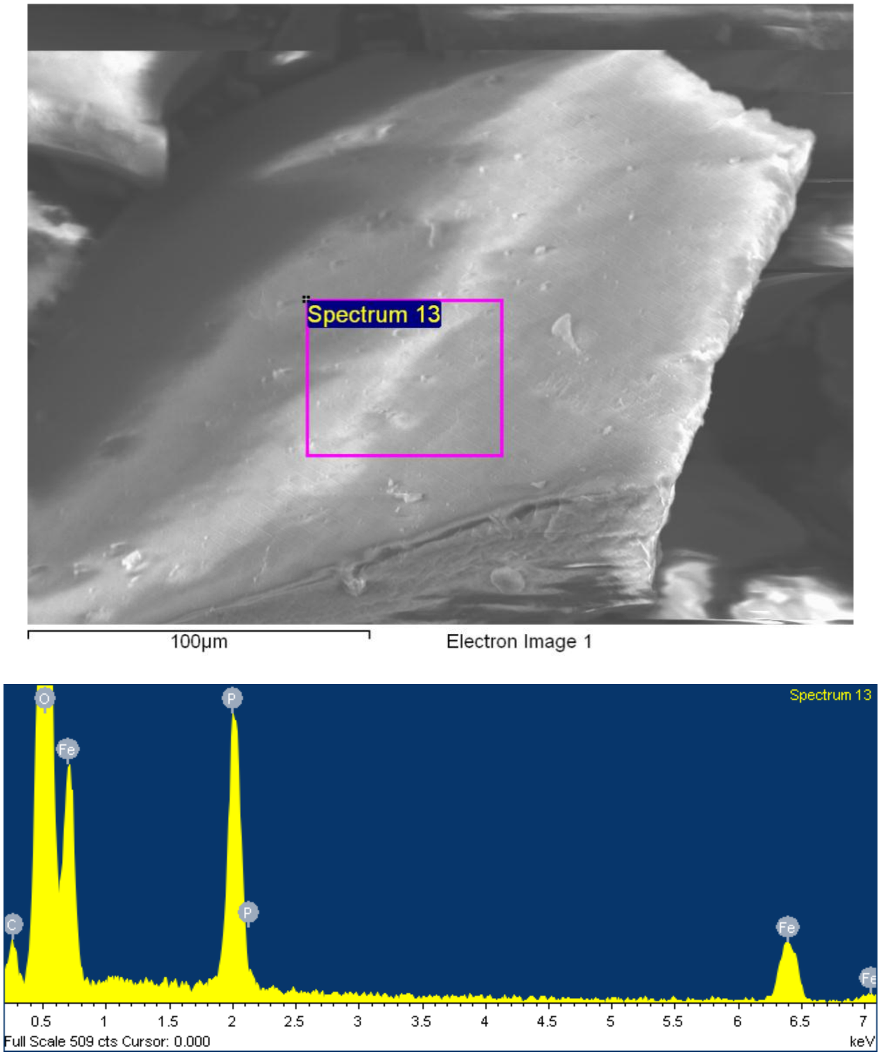

Figure 1: SEM and EDS analysis of precipitated particles filtered from acid solution for mixed $LiFePO_4$ and $LiCoO_2$

Next our method was tested to see if it could, separate Fe from Mn: 0.2355g of $LiFePO_4$ and 0.2635g of $LiMn_2O_4$ were mixed in 20mL of 4M H_2SO_4 and 4mL of H_2O_2 at 74℃ for 2hrs. Undissolved $LiFePO_4$ was filtered from solution and NaOH was added to the solution until the pH=3.0. SEM and EDS results (Figure 2) show that no Mn existed in $Fe(OH)_3$ precipitate. 0.2ppm of iron was detected in the solution by AAS due to the lower pH. Therefore, the Mn was mostly separated from Fe but the separation should improve when the pH is increased to precipitate more $Fe(OH)_3$.

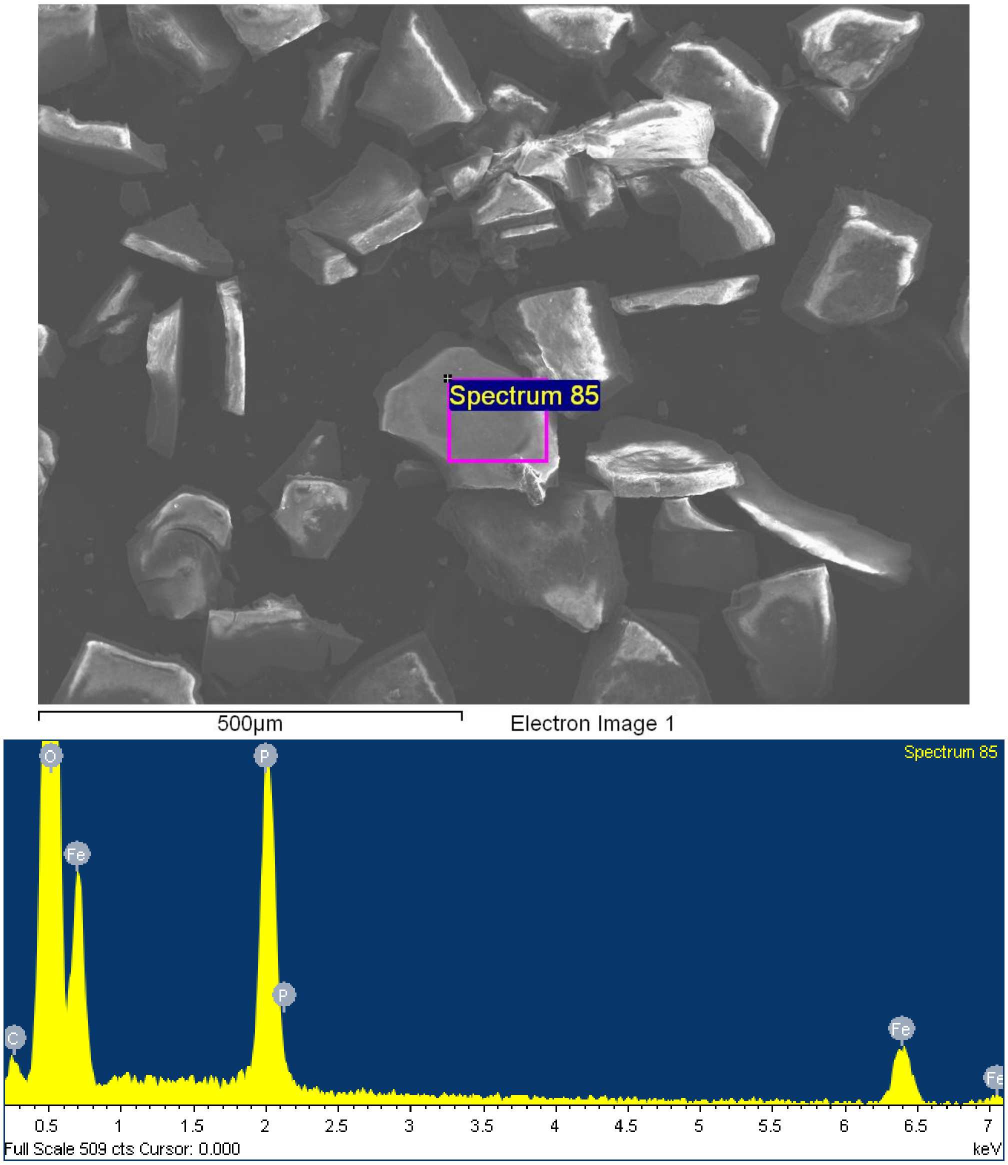

Figure 2: SEM and EDS analysis for precipitated particles filtered from acid solution for mixed $LiFePO_4$ and $LiMn_2O_4$

Similarly, our method of separating Fe from Ni, Mn and Co was analyzed: 0.2350g of $LiFePO_4$ and 0.2759g of $LiNi_{0.33}Mn_{0.33}Co_{0.33}O_2$ were mixed in 20mL of 4M H_2SO_4 and 4mL of H_2O_2 at 74℃ for 2hrs. Undissolved $LiFePO_4$ was filtered from solution and NaOH was added to the solution until the pH=3.22. $LiNi_{1/3}Mn_{1/3}Co_{1/3}O_2$ was dissolved in a solution of sulfuric acid and hydrogen peroxide. It is believed that Mn, Co and Ni existed as divalent metal ions in the solution. AAS test showed that the concentration of Fe ions left in solution was 0.162ppm. SEM and EDS results (Figure 3) showed that $Fe(OH)_3$ precipitate filtered from the solution did not contain Co, Ni or Mn.

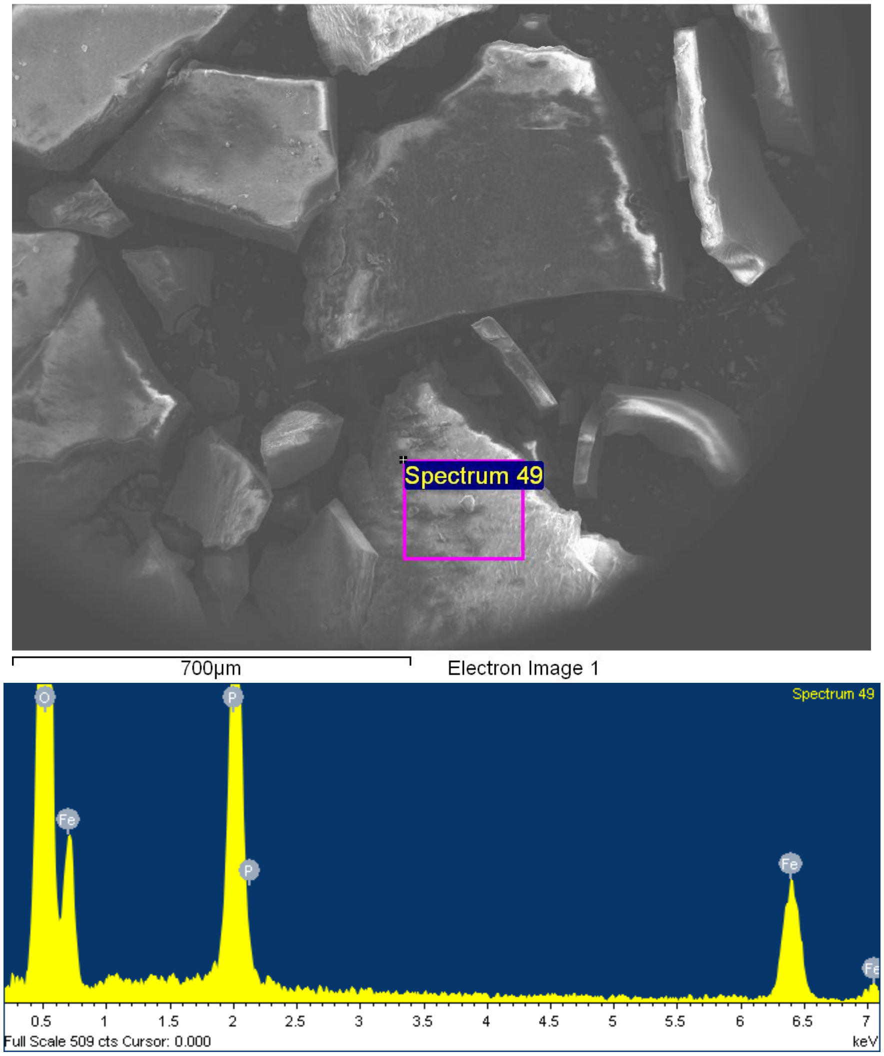

Figure 3: SEM and EDS analysis for precipitated particles filtered from acid solution for mixed $LiFePO_4$ and $LiNi_{0.33}Mn_{0.33}Co_{0.33}O_2$

Our method of separating Fe from Ni, Mn and Co was tested again this time: using 0.083g $LiFePO_4$, 0.0996g$LiCoO_2$, 0.1074g $LiNi_{0.33}Mn_{0.33}Co_{0.33}O_2$, and 0.1021g $LiMn_2O_4$. The materials were mixed in 20mL of 4M H_2SO_4 and 4mL of H_2O_2 at about 74℃ for 2hrs. This time the pH was increased to3.43. The concentration of Fe ions left in the solution was measured with AAS to be 0.041ppm. EDS results (Figure 4) showed that $Fe(OH)_3$ precipitate filtered from the solution did not contain Co, Ni or Mn. Therefore, by adjusting the pH, we can successfully separate Fe from Co, Ni and Mn in the solution, which will help us co-precipitate a mixture of $Co(OH)_2$, $Ni(OH)_2$ and $Mn(OH)_2$ in the next step.

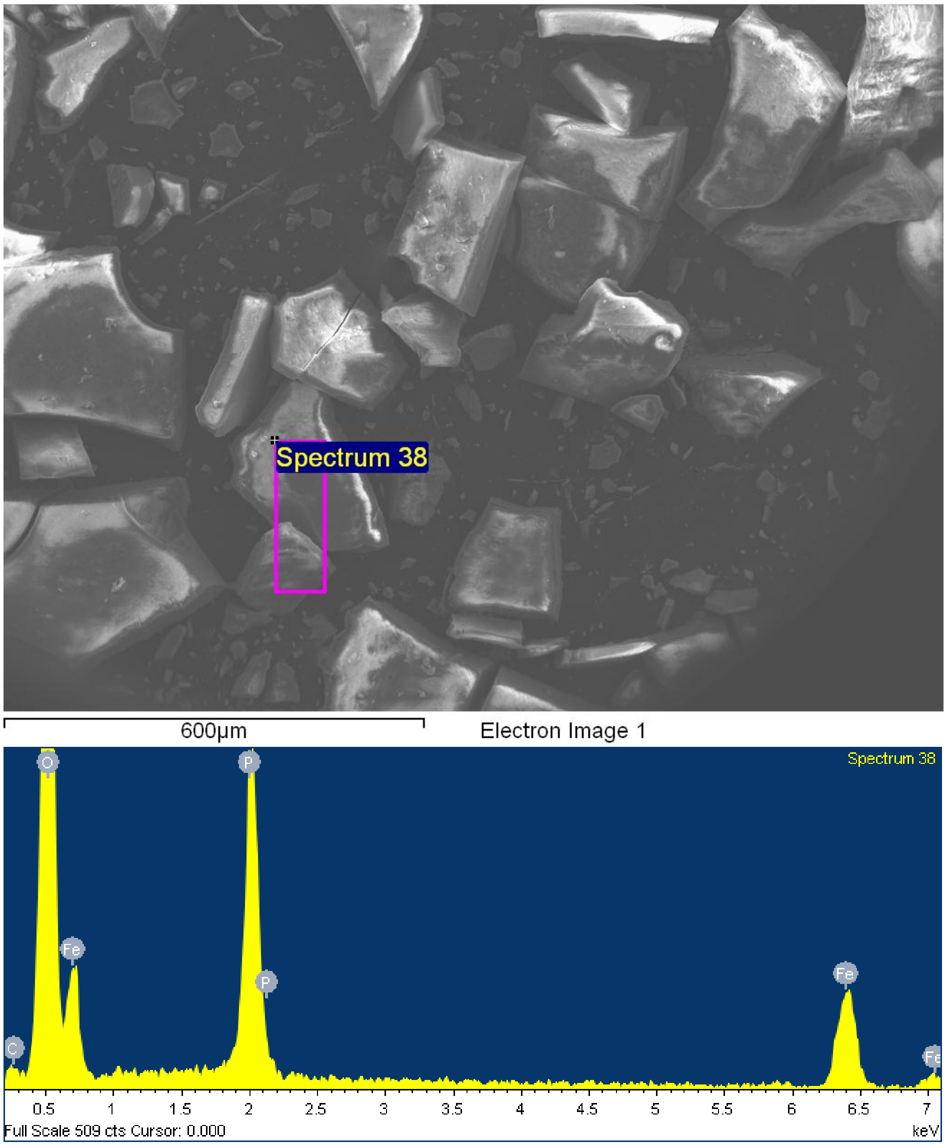

Figure 4: SEM and EDS analysis for precipitated particles filtered from acid solution for mixed $LiFePO_4$, $LiCoO_2$, $LiMn_2O_4$ and $LiNi_{0.33}Mn_{0.33}Co_{0.33}O_2$

Processing spent Li-ion batteries

The procedure described in this paper regenerates new cathode materials from other (potentially used) cathode materials. In an industrial process the Li-ion batteries must be processed to a point at which cathode materials are separated from the other battery components and can be leached into solution. This is typically done by shredding or crushing the batteries. In order to shred Li-ion batteries the batteries must be discharged or shredded in an inert atmosphere, to prevent organic electrolyte from catching fire. After shredding the steel casing is removed by magnetic separation. The electrolyte can be recovered, if desired, by solvent extraction in a reusable organic liquid. Then the cathode materials must be separated from the aluminium current collector, this has been accomplished by dissolving the aluminium in basic solution, dissolving the binder in NMP and physical processes such as ball milling or crushing The battery processing approach is shown in Figure 5. The chemical cost of these process is relatively low since any organics used can be reused and sodium hydroxide is relatively cheap requiring approximately $30/ton of batteries to dissolve all of the aluminium.

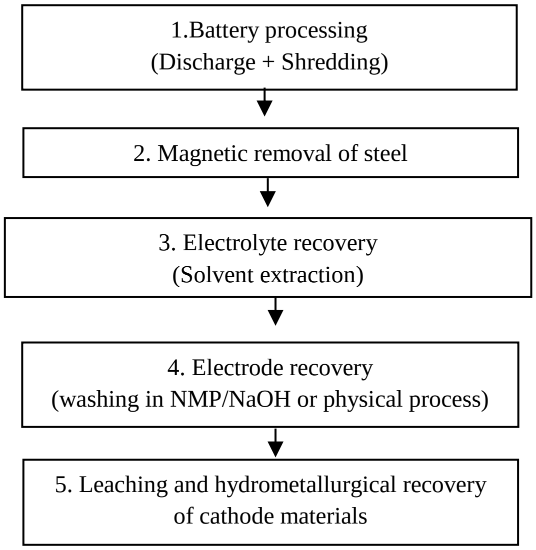

Figure 5: Li-ion battery processing procedure