Impacts of solvent washing on the electrochemical remediation of commercial end-of-life cathodes

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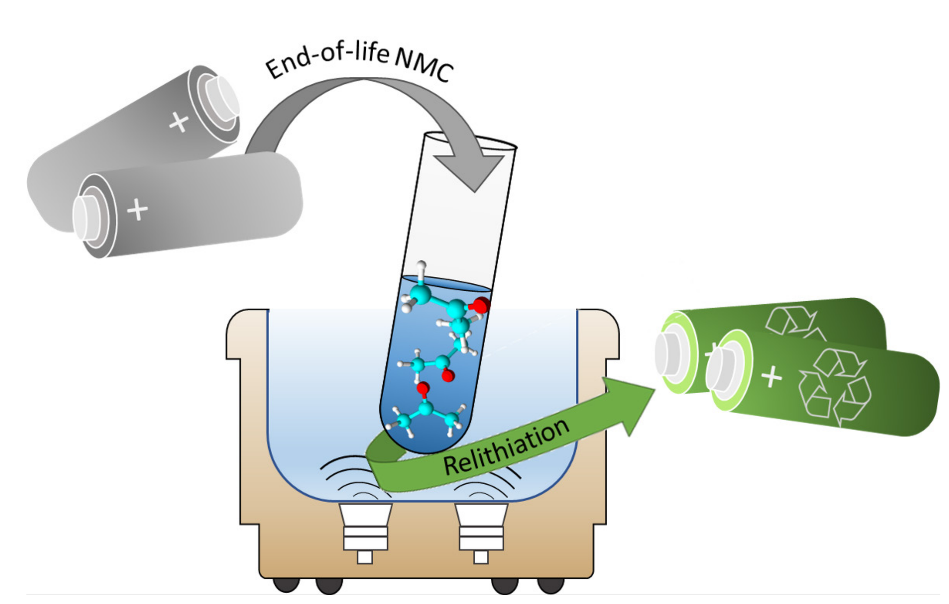
NREL ReCell ADVANCED BATTERY RECYCLING

Background



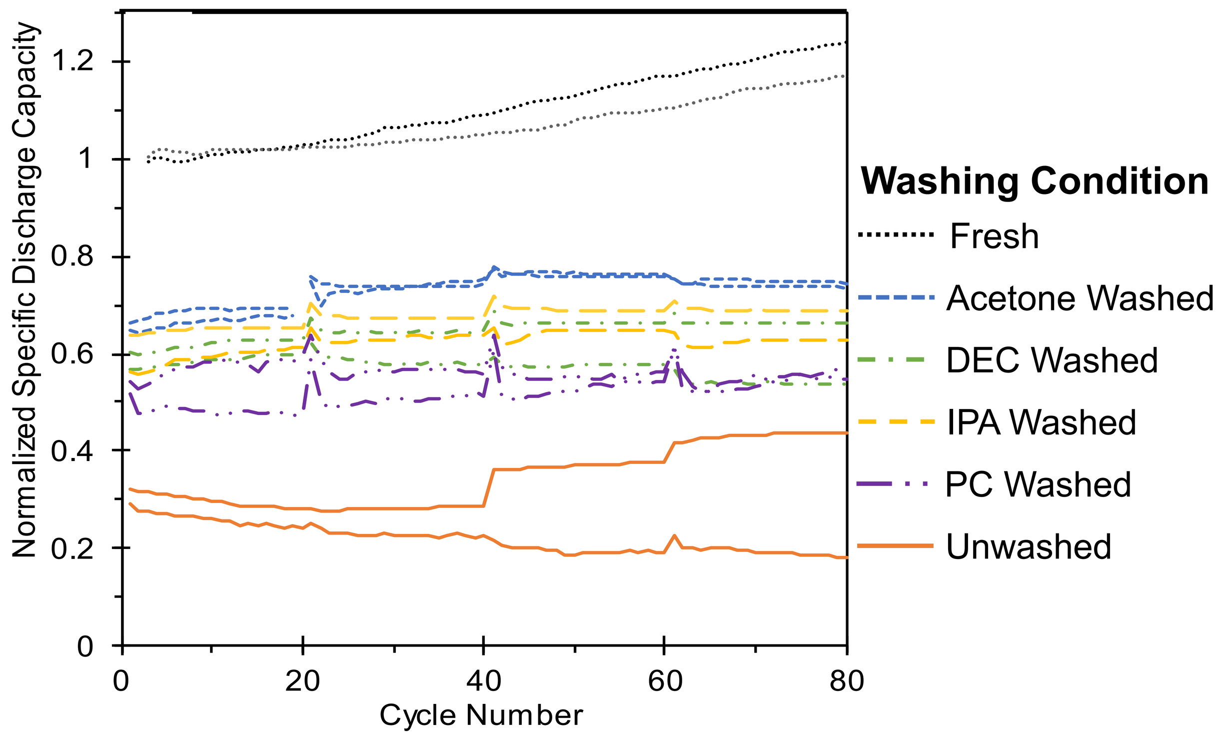
- Novel direct recycling techniques aim to rejuvenate battery cathode materials through the reintroduction of Li^+ ("relithiation").
- The effectiveness of relithiation is impacted by end-of-life cathode surface chemistry.
- Rational optimization of pre-treatment methods can improve the efficacy of subsequent remediation strategies.

Objectives

- Determine the role of varying physicochemical solvent properties on the mechanism of capacity recovery in relithiated cathodes.
- Identify target solvent properties to optimize a practical washing protocol in the context of battery direct recycling.

Highlights

- Up to 174% capacity recovery is achieved for acetone-washed cathode material.
- Highly nucleophilic solvents are found to favor enhanced cathode recovery.
- Nucleophilic solvents improve relithiation efficacy through:
  - -Reduced charge-transfer impedance
  - -Reintroduction of bulk lattice oxygen
  - -Selective removal of surface species



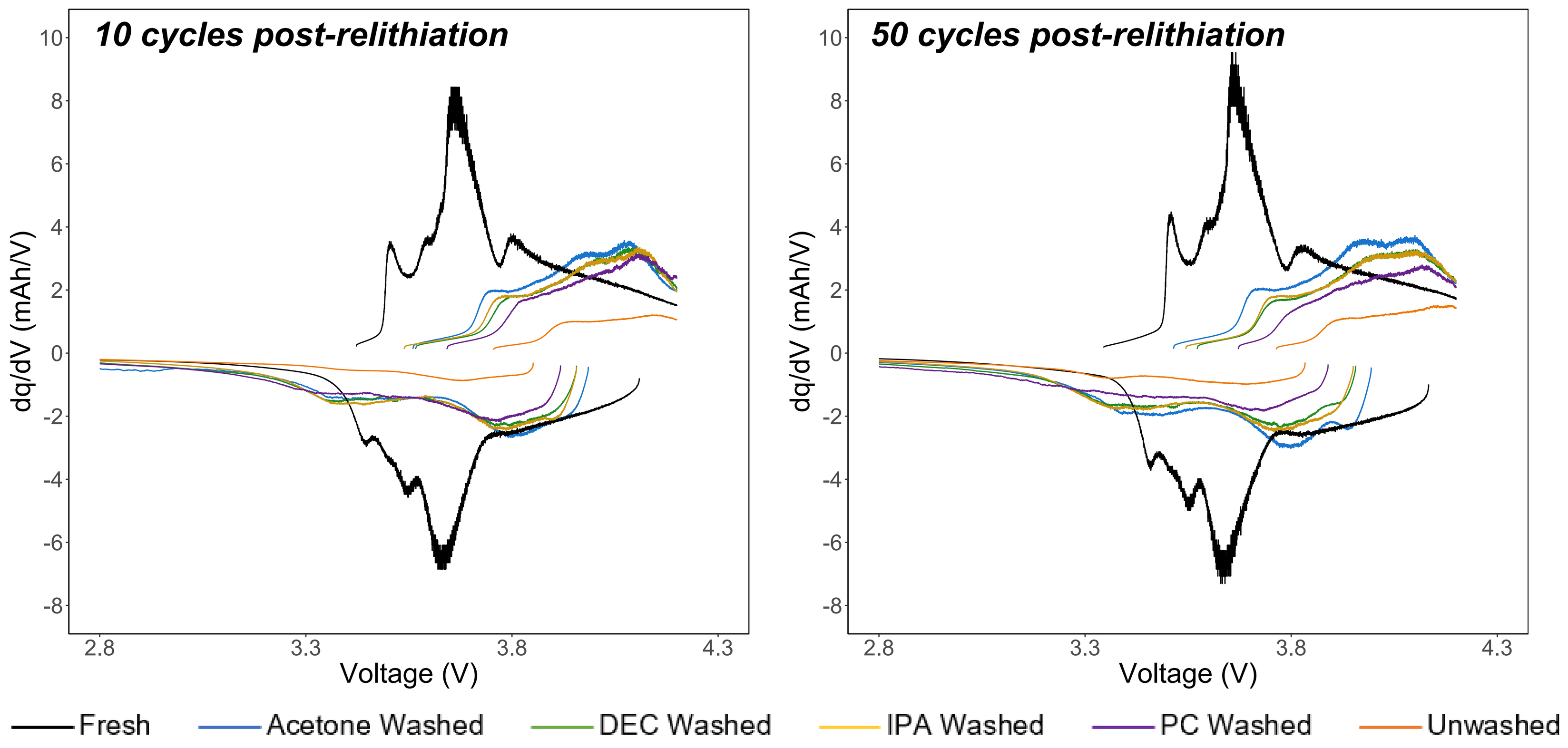
Electrochemical full-cell cycling performance of relithiated cathode material, with and without solvent washing.

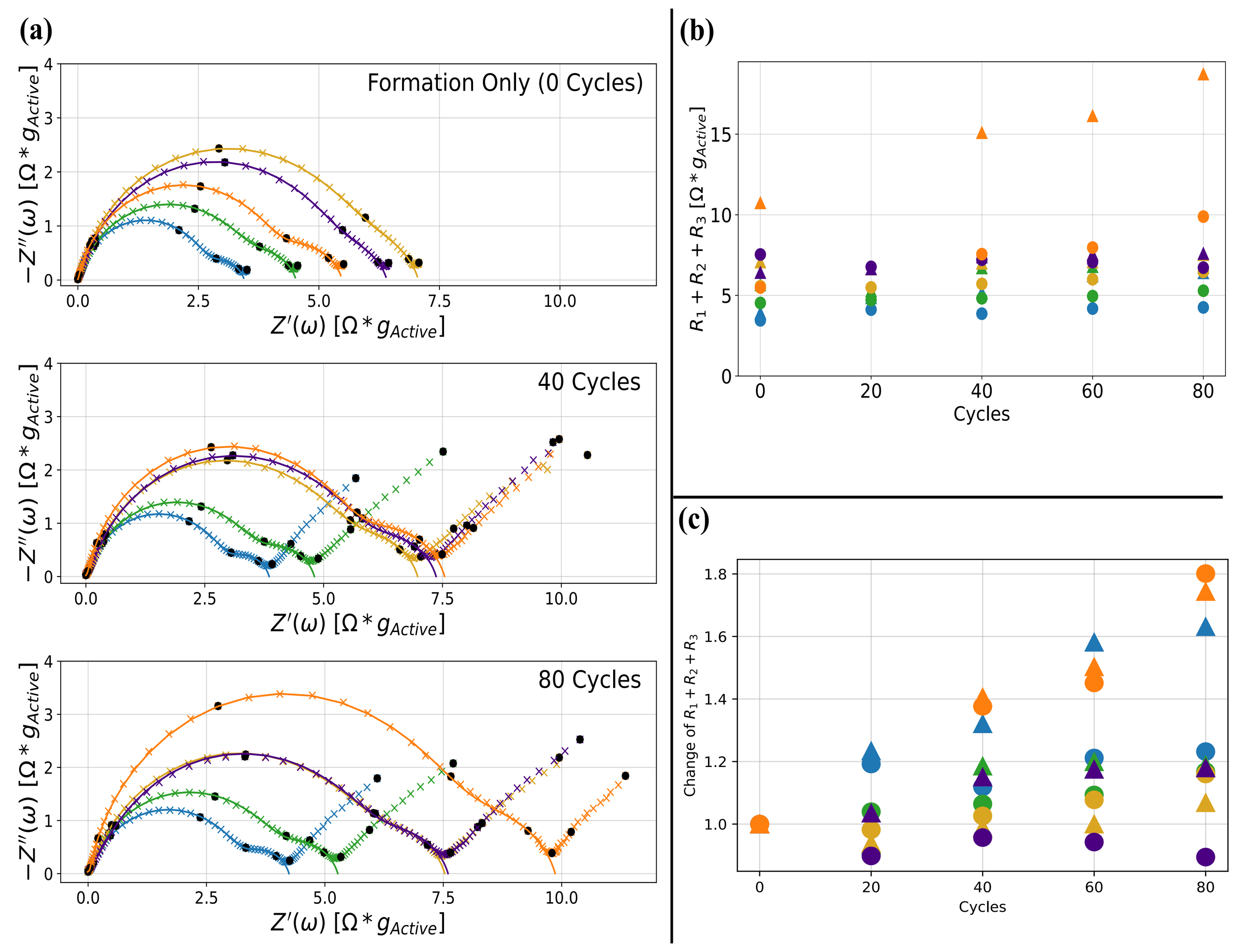
Approach

- Cathode material was recovered from heavily aged commercial 40 Ah pouch cells ($Li(Ni_{0.41}Mn_{0.36}Co_{0.23})O_2$ vs graphite).
- Cathode black mass (NMC + binder + conductive carbon) was sonicated with each of four solvents: **acetone, diethyl carbonate (DEC), isopropyl alcohol (IPA), propylene carbonate (PC)**.
- Treated and untreated black mass was recast and fabricated into electrochemical half cells (NMC vs Li^0).
- Electrochemical relithiation was conducted in half cells; cathodes were recovered and reassembled in full cells.
- Electrochemical (**galvanostatic cycling**, **dQ/dV**, **EIS**), structural (**XRD**), and chemical (**GC-MS**) characterization was conducted on electrodes, treated powders, and wash solutions, respectively to probe mechanisms of reactivity.

Results

Electrochemical Analysis

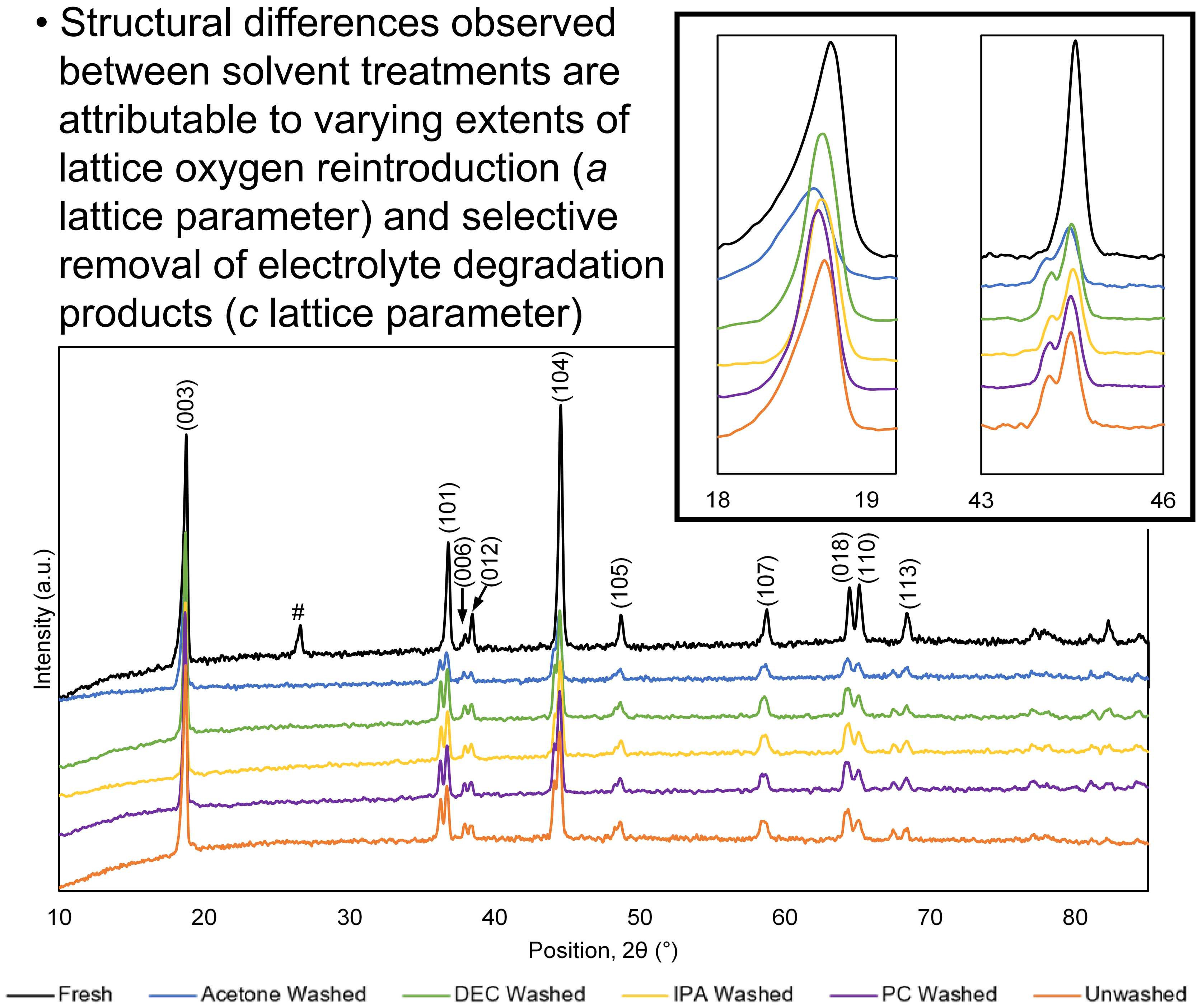




- Capacity recovery for washed + relithiated samples is tied to anionic redox, rather than recovery of transition-metal redox. Highly nucleophilic solvents (acetone) reduce the NMC surface, favoring reintroduction of bulk lattice oxygen.
- All washing conditions reduce overall impedance; acetone and DEC washes most improve charge-transfer resistance.

Structural Analysis

- Structural differences observed between solvent treatments are attributable to varying extents of lattice oxygen reintroduction (*a* lattice parameter) and selective removal of electrolyte degradation products (*c* lattice parameter)



Chemical Analysis

- Species removed by solvent washing include electrolyte degradation products, additives, and electrolyte-additive reaction products.
- Polarity, nucleophilicity, and sterics of the solvent impact the selectivity of surface species removal from spent cathodes.
- Acetone (ketone) effectively removes all three classes of end-of-life compounds.

Summary & Outlook

- Mechanism of capacity recovery for solvent-washed, relithiated cathodes includes both surface purification and surface reconstruction.
- High nucleophilicity and moderate polarity are found to promote improved relithiation.
- Optimization at scale should include further solvent tailoring, TEA, and safety analysis.

For further details:
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