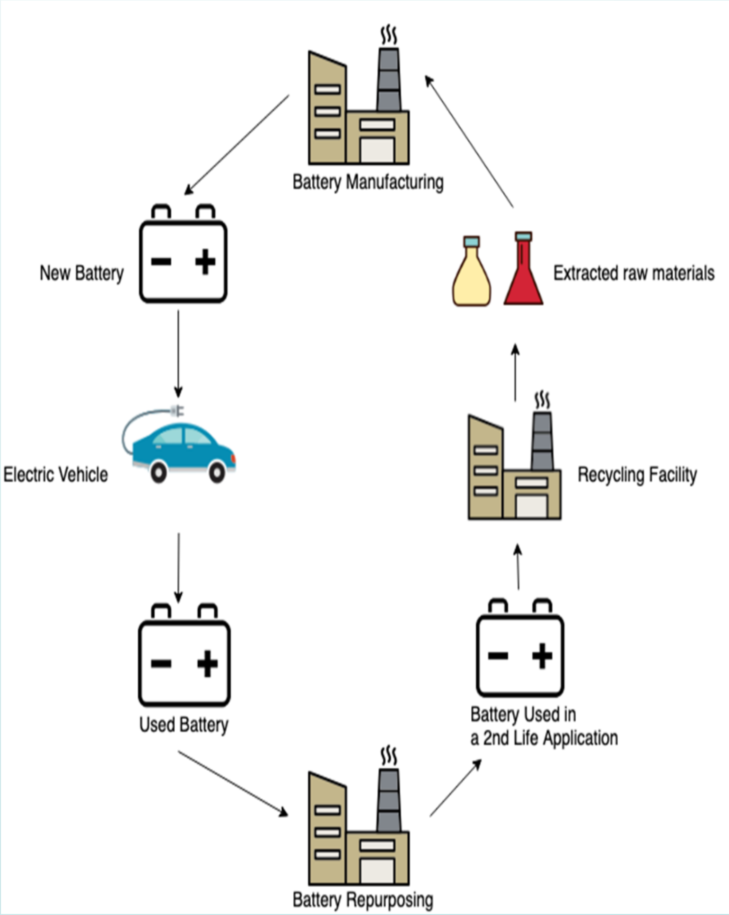
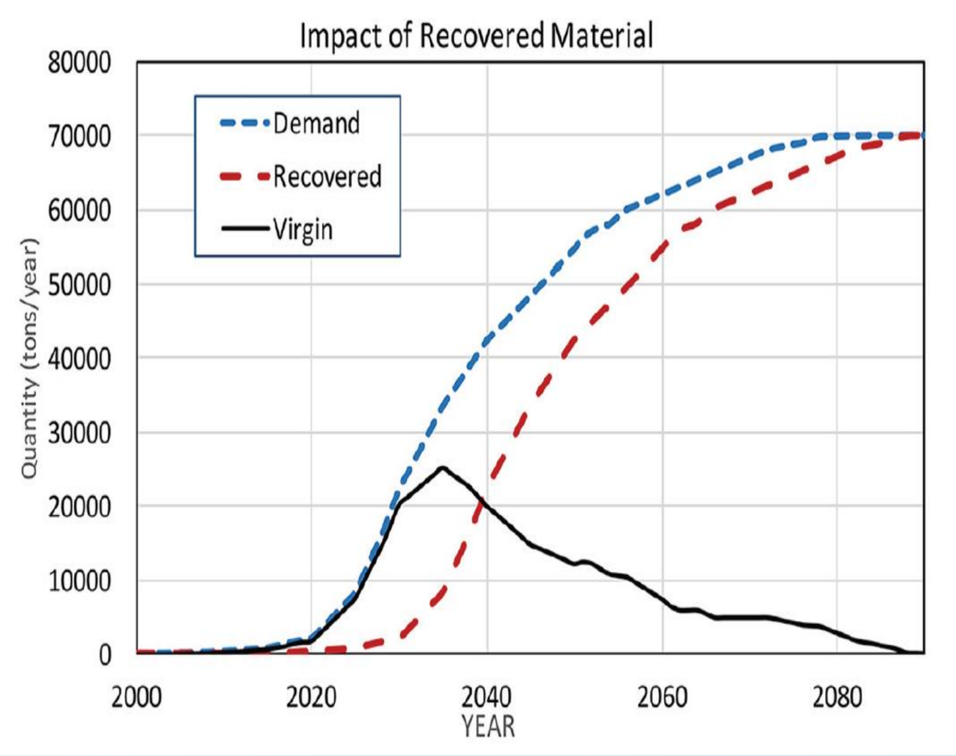
Li ion battery durability vs. a circular economy strategy

The battery value chain in a circular economy

- Global sustainability trend to encourage strategies supporting a circular economy for Li ion batteries
  - China battery recycling regulation
  - EU – The Circular Economy Action Plan / The Green Deal
  - World Economic Forum – Global Battery Alliance
- A circular battery value chain assumes
  - Extended life time of already existing batteries (repair and reuse)
  - Substances and materials will be recoverd and reused for manufacturing of new batteries in an endless closed loop



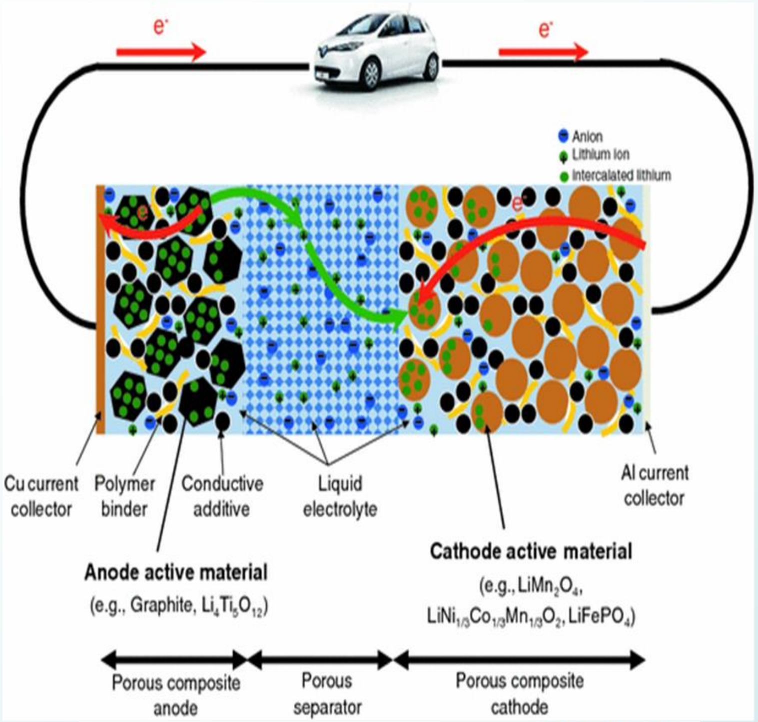
Projected Li ion battery material availability

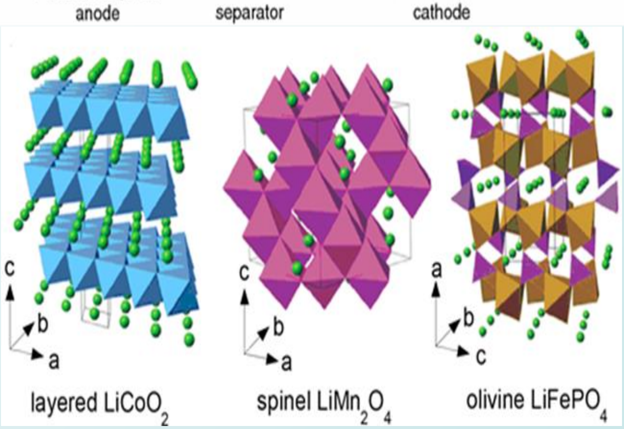


Source: L. Gaines (2018) Argonne National Labs. Key issues for Li battery recycling

- The demand for battery materials will continue to increase exponentially in a foreseeable future
- Availability of recovered materials for Li ion traction batteries will begin to increase approx 2030, but still at a modest scale compared to demand
- With existing regulatory strategies, recycled materials can become a significant portion of materials going into cell manufacturing post 2040

Review of Li ion cell chemistry





- The objective of the electrochemical cell is transform chemical energy into electrical energy (discharge) and vice versa (charge)
- Li ion cell chemistry is an intercalation process
- Reaction at discharge

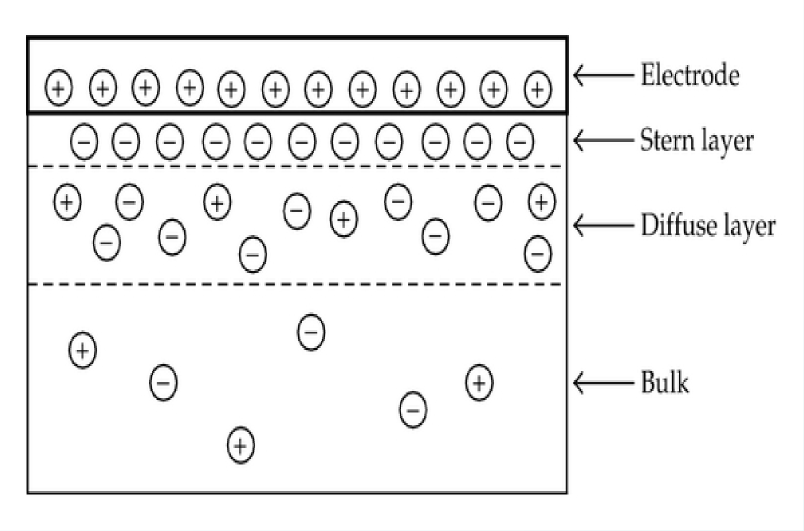
  Cathode = reduction:

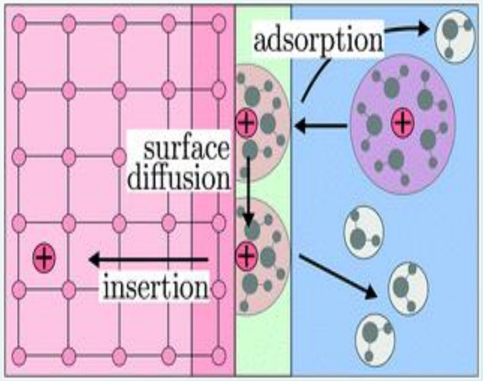
  $MO_2 + Li^+ + e^- \rightarrow LiMO_2$

  Anode = oxidation:

  $LiC_6 \rightarrow Li^+ + e^- + C_6$

Electrode reactions





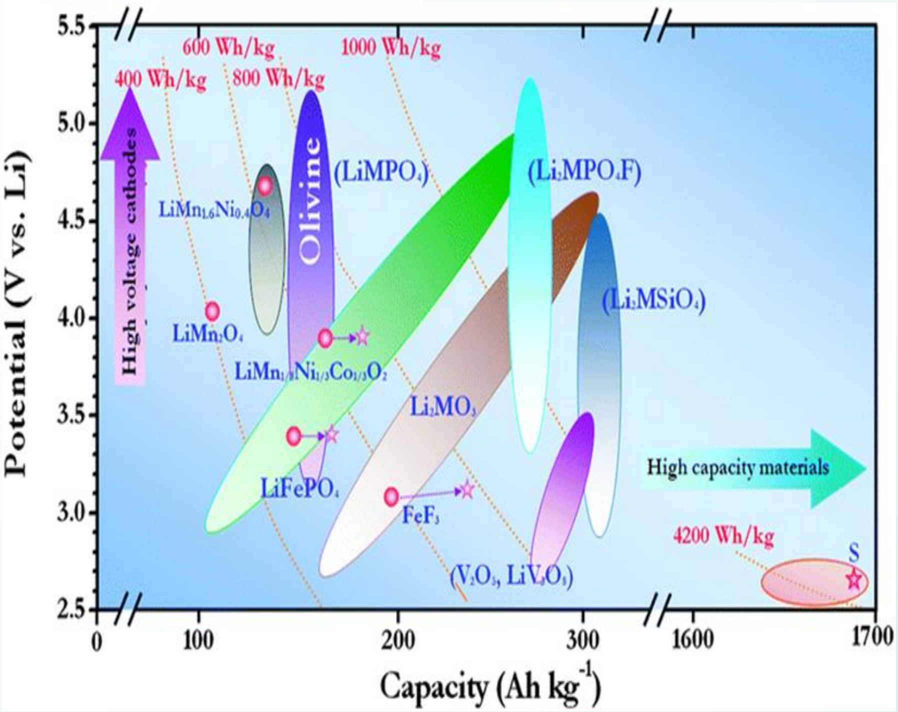
Intercalation process at the cathode-electrolyte interface

- Electrode reaction occur at the interface between electrode and electrolyte and comprises multiple steps, e.g.
  - Diffusion of ions to the electrode surface
  - Adsorption
  - Surface diffusion
  - Charge transfer
- Intercalation involves an insertion step when the Li ion takes place in the electrode material's crystal lattice
- Electrode reactions are strongly dependent on the chemical composistion and the physical characteristics of the electrode surface
  - Reaction rates
  - Overvoltage (energy losses)
- Crystal defects affect Li ion mobility in the electrode material

Adsorption: $Li^{+} + S + (1-\delta)e^{-} \rightarrow LiS^{\delta+}$

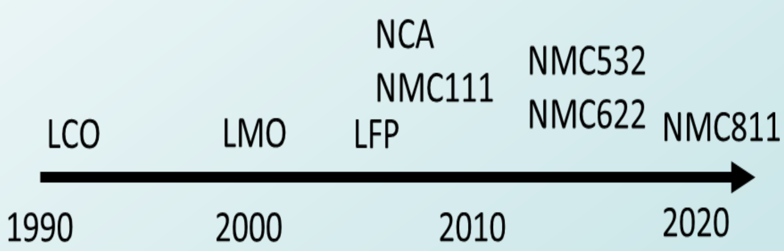
Intercalation: $LiS^{\delta+} \rightarrow Li^{+}(s) + S + (1-\delta)e^{-}$

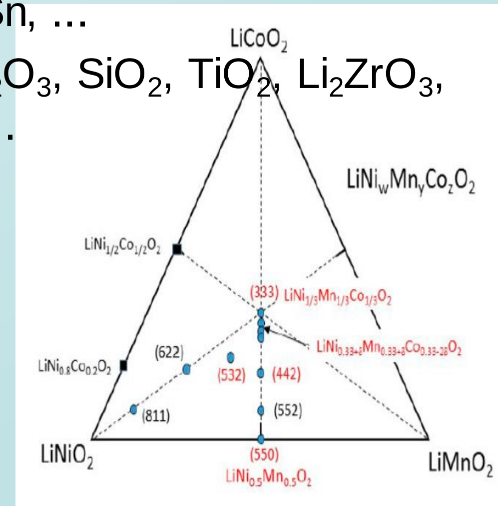
Cathode materials



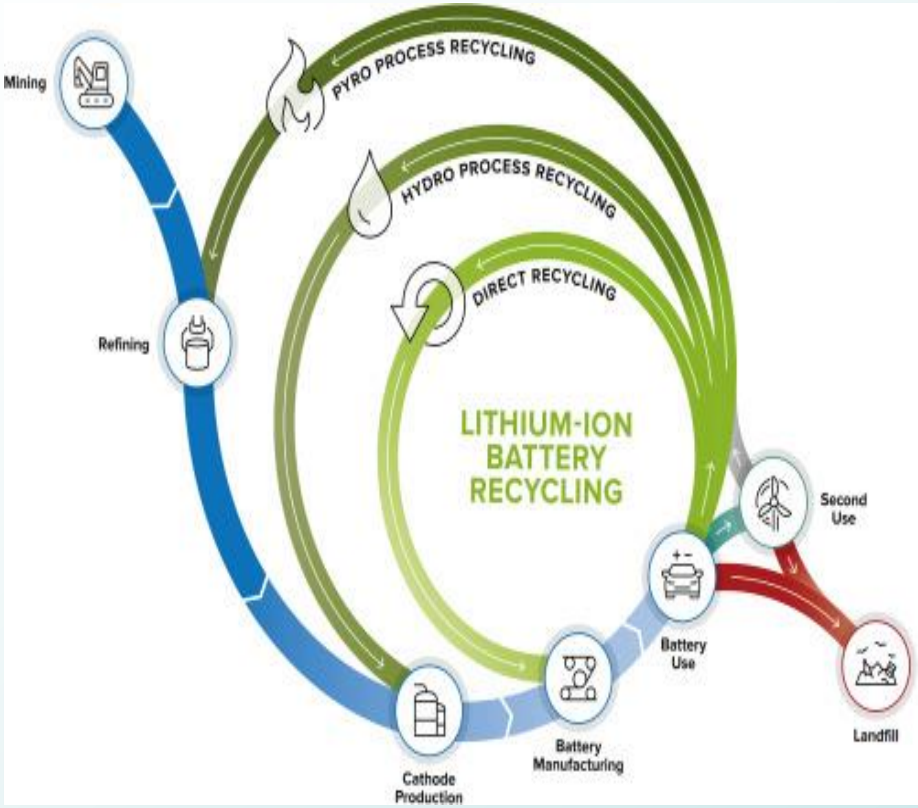
- Predominantly transition metal oxides and phosfates
  - Layered: $LiCoO_2$ (LCO), $Li(Ni_xMn_yCo_{1-x-y})O_2$ (NMC) $LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$ (NCA)
  - Spinelles: $LiMn_2O_4$ (LMO)
  - Olivines: $LiFePO_4$ (LFP)
- ”Invisible ” components in the form of dopic agents and coatings
  - Dopning: Al, Fe, Mo, Ru, Zn, La, V, Cr, Mg, Zr, Sn, ...
  - Coatings: Al_2O_3, SiO_2, TiO_2, Li_2ZrO_3, V_2O_5, AlF_3, ...

Katodmaterial i kommersiella celler:





Recycling of Li ion batteries - Overview



- 3 main recycling processes:
  - Pyrometalurgic process
    - Treats battery waste as ”ore” and feeds into the refining step
    - Recovers Co, Ni and Cu
  - Hydrometallurgic process
    - Metal components are recovered by leaching following an initial separation and grinding step
    - The multi-step leaching process involving cyclic extraction using strong acids, complex binders and 2 phase separation
    - Recoverd materials: Al och Cu foils (current collectors), Co, Ni, Mn and Li from cathode
    - Recovered substances are used as precursors to electroce materials
  - Direct recycling
    - Process is under development
    - All materials can be recovered
    - The materials are recovered as close to their final engineering/manufacting state as possible
    - Customized to specific cell chemistries/compositions
    - Economically justified för Co-poor chemistries

Source: L. Gaines (2019) Argonne National Labs. Profitable Recycling of Low-Cobalt Lithium-Ion Batteries Will Depend on New Process Developments

Research status: Li ion battery material recovery and use of secondary raw materials

- Relatively new research area
- Currently the focus of activities is centered in China and USA but rapidly growing in EU, based on scientific publications
- Existing studies are based on laboratory studies involving very well defined ”waste streams”, i.e. single cell model from one manufacturer
- Ageing of original cells prior to recycling is reportedly performed in laboratory environment
- Preparation of ”waste material” is performed manually
- Recovered secondary raw materials have a different contamination profile compared to pristine raw materials
- Although recovery rates are reported, there is little or no information on the quality of the recovered secondary raw materials compared to pristine raw materials
- General experience is that high recovery rates usually are connected with a lower quality of the recovered materials, due to co-extraction of metals
- Manufacturing of high performance Li ion batteries relies on use of high quality, battery grade materials
- With the exception of Pb-acid batteries, the general experience is that only limited amounts of secondary raw materials can be used to manufacture new cells without introducing performance penalties
  - Rechargeable NiCd and NiMH less than 30%
  - For primary alkaline less than10%
  - There is no evidence supporting that the performance of Li ion batteries would not be equally affected.

Challenges with recovered materials

Substance recycling

- Substance contaminants are expected in the recovered material:
  - Metallic contaminants: Cu, Al, Fe
  - Non-metallic contaminants: C, F, P
- Al and Fe can interfere with hydrometallurgic metal extraction
- Small amounts of contaminants can result in deteriorated electrochemical properties
  - Cu contamination is common in recycled Co – degrades energy density
  - Cu contamination also commin in Ni due to co-extraction
  - Al contamination contributes to increased nucleation, resulting in smaller but more numerous particles
  - Al contamination >0.6% contributes to increased rate of capacity fade (6-14% during 500 cycles)
- Material degradation compared to pristine materials due to accumulation of different contaminants in the end product

Direct recycling

- Due to the continual changes in Li ion chemistries, the recovered materials are likely to be obsolete and require additional engineering to be suitable for use
- Relies on strictly homogeneous waste streams (chemistry and manufacturer specific)
- The material morphology degrades during recycling
  - Uneven particles
  - Structural effects on both primary and secundary particles
  - Lower ”tap density” resulting in lower energy density
- Risk of inhomogeneous coatings on particles and surface contaminants
- Successive increased degradation with multiple useage-recycling cycles
- Effectiveness of the recycling process is linked to SOH of waste material
- High sensitivity to Al and Cu microparticle contamination which degrades electrical performance

Summary

- The consequences of an increased use of recycled materials in Li ion production on electric performance including durability, is difficult to foresee
  - A complex waste stream makes it challenging to recover substances and/or materials with high quality
  - Current research indicates that adverse effects on battery cycle life can be expected due to contamination and morphology effects
- The assumption of continued (linear) increase in Li ion traction battery energy density and cycle life may not be valid in a circular battery economy
- The Battery Durability GTR cannot be developed isolated from other battery sustainability regulation
  - Needs flexibility to reflect battery technology development:
    - New cell chemistries/technologies
    - Performance impacts of material recycling in cell manufacturing
- More research is needed to understand how different sustainability regulation and requirements may impact on battery durability performance and, hence, realistic performance targets for the Battery Durability GTR