

Leveraging Reversible Chemistry for Materials Sustainability

in Energy Storage

Zheng Chen, Ph.D.

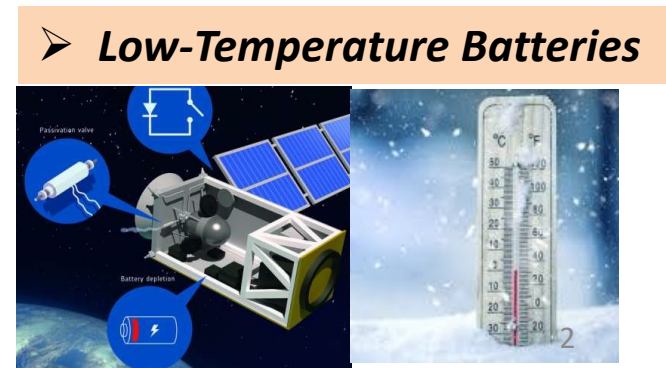
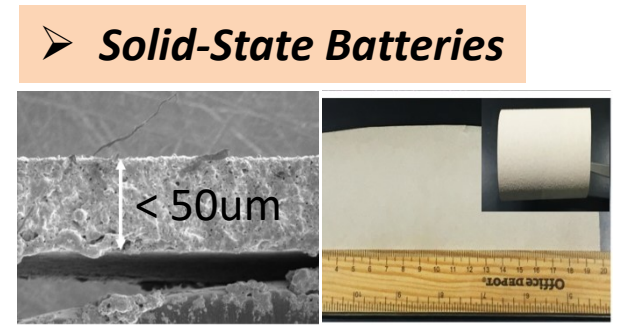
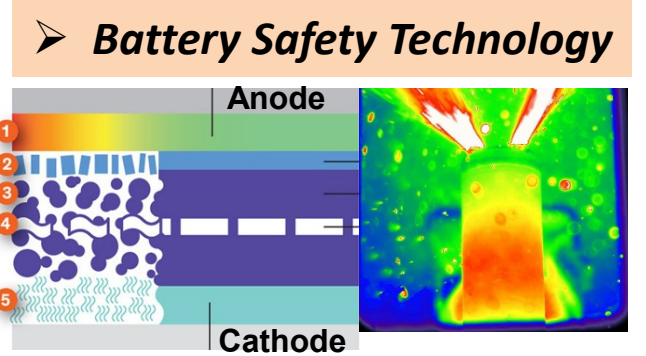
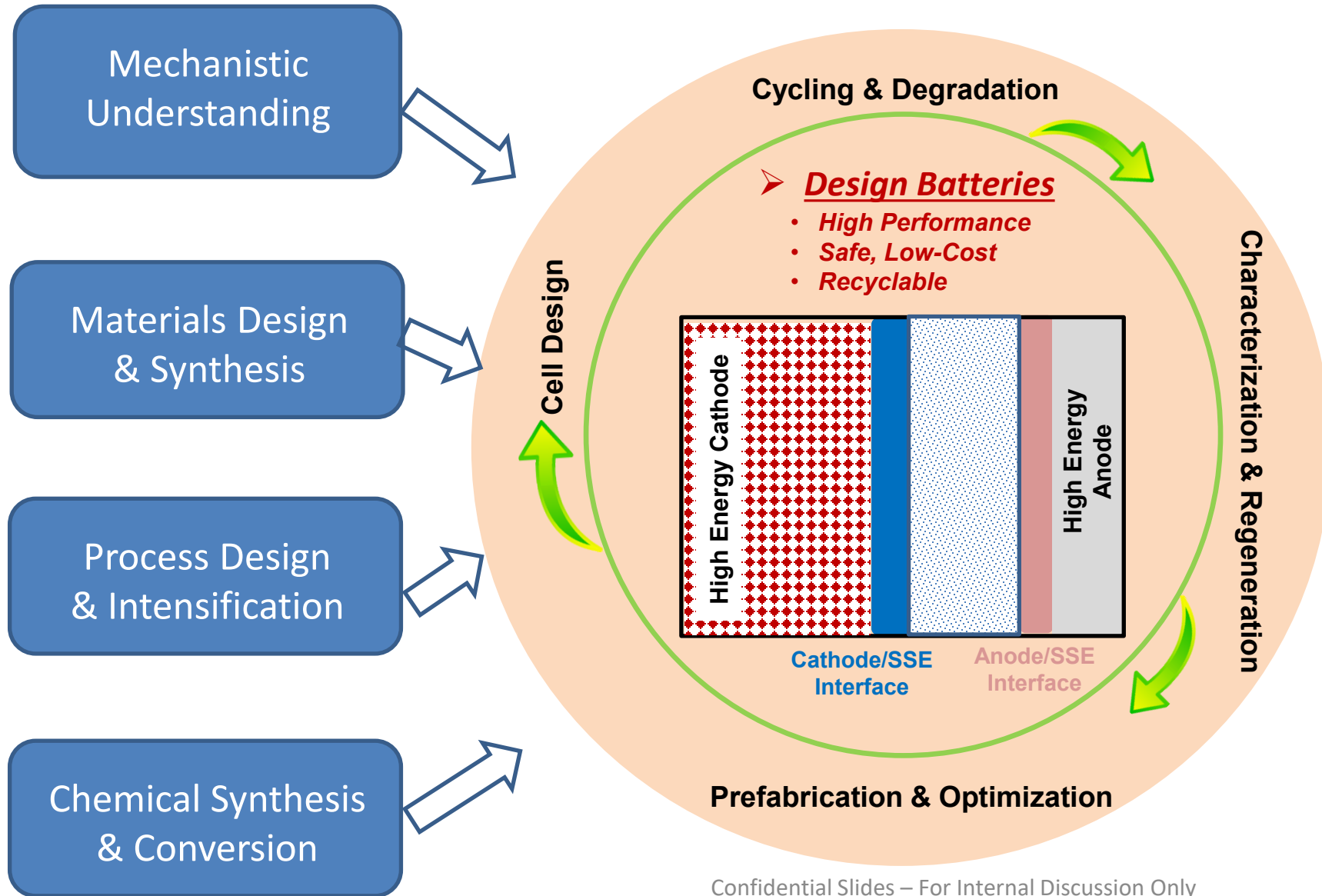
Assistant Professor, Department of NanoEngineering/Chemical Engineering
Sustainable Power and Energy Center (SPEC)

University of California, San Diego

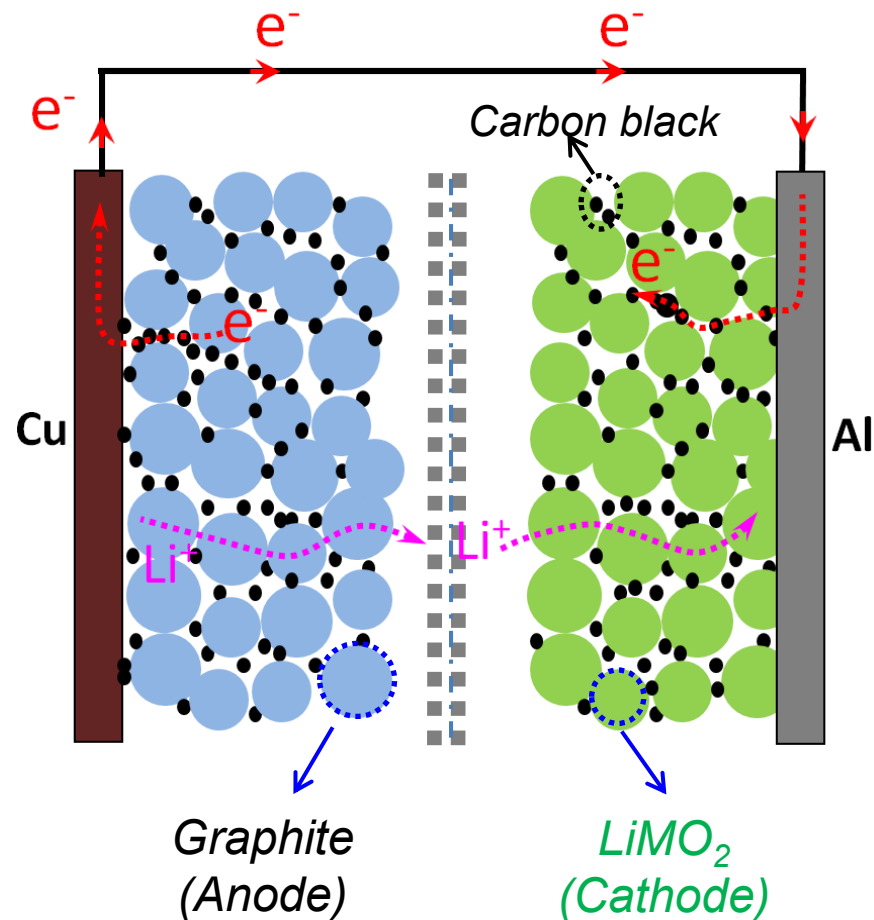
E-mail: zhengchen@eng.ucsd.edu



Materials Science and Chemical Technology for Sustainable Energy



Critical Materials Are Heavily Used in Li-ion Batteries



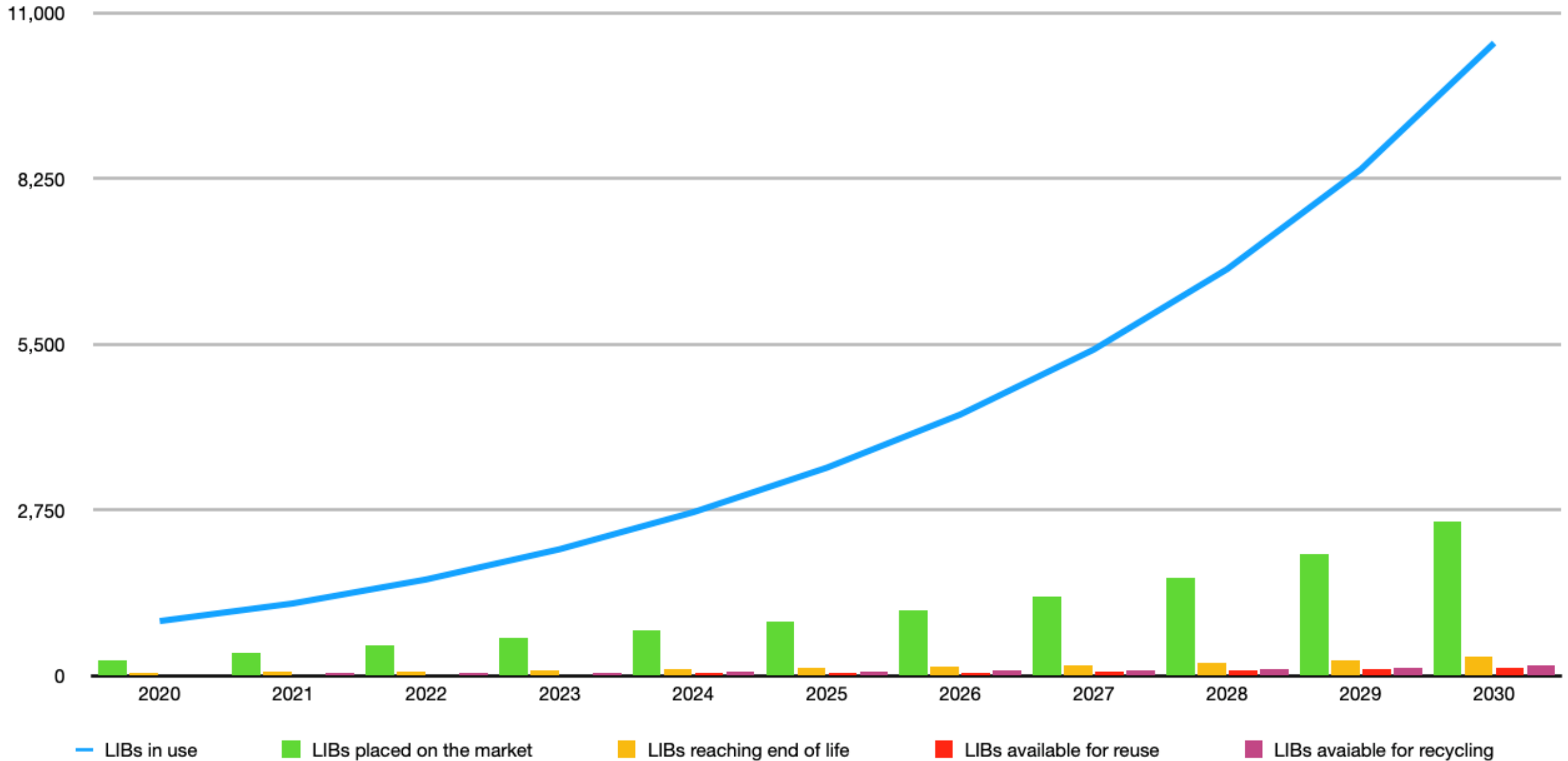
Cathode Materials:

- LiFePO_4
- LiMn_2O_4
- LiCoO_2
- $\text{Li}_x\text{Ni}_y\text{Mn}_z\text{Co}_{1-y-z}\text{O}_2$
(NMC111, NMC532...)
- $\text{LiNi}_{0.8}\text{Co}_{0.15}\text{Al}_{0.05}\text{O}_2$

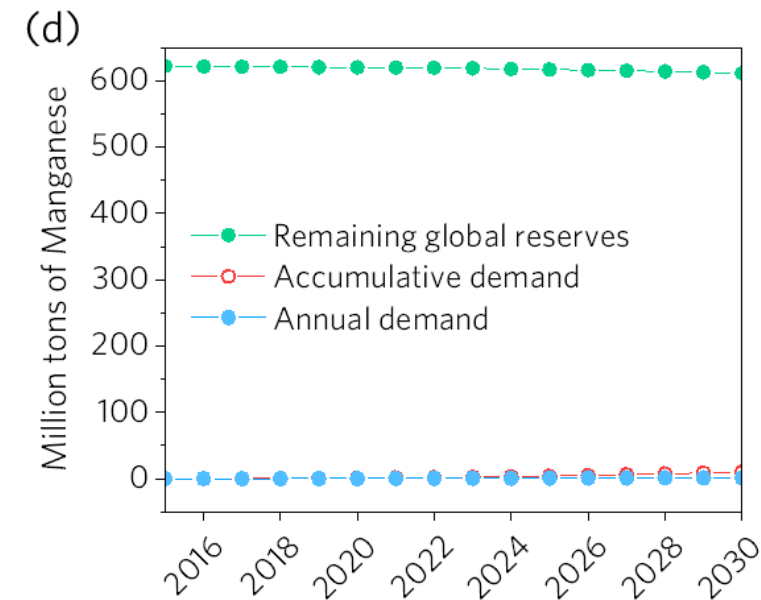
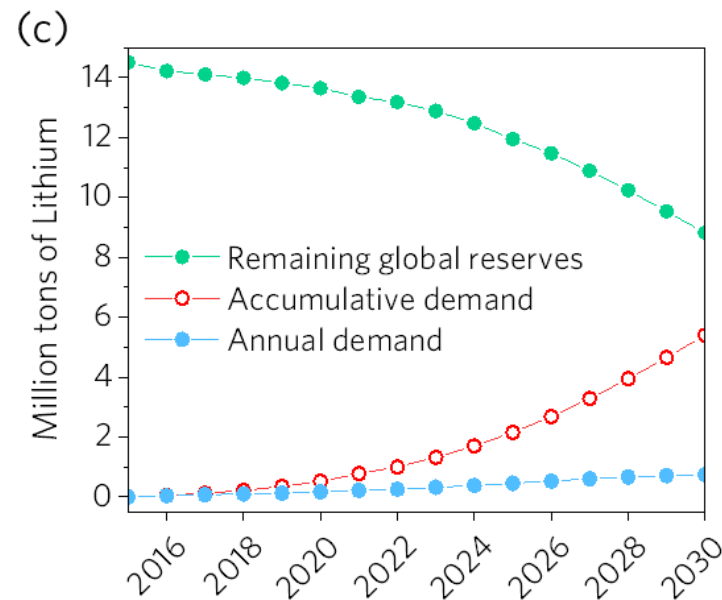
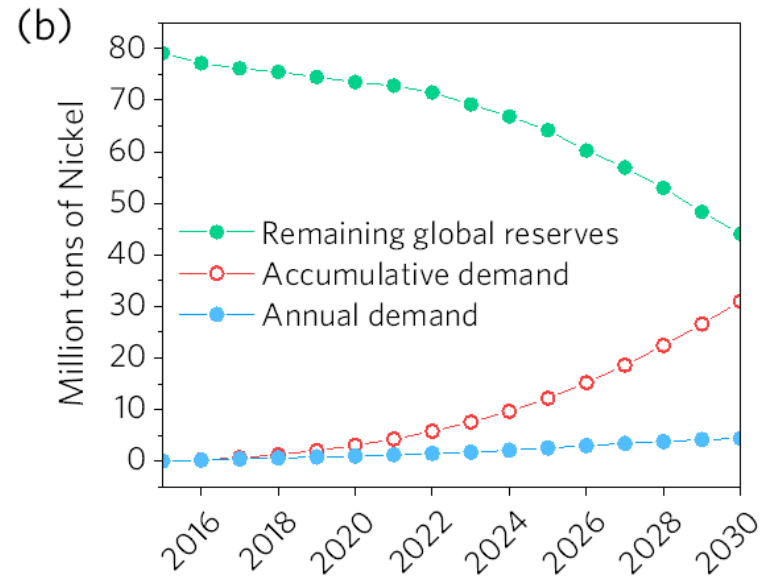
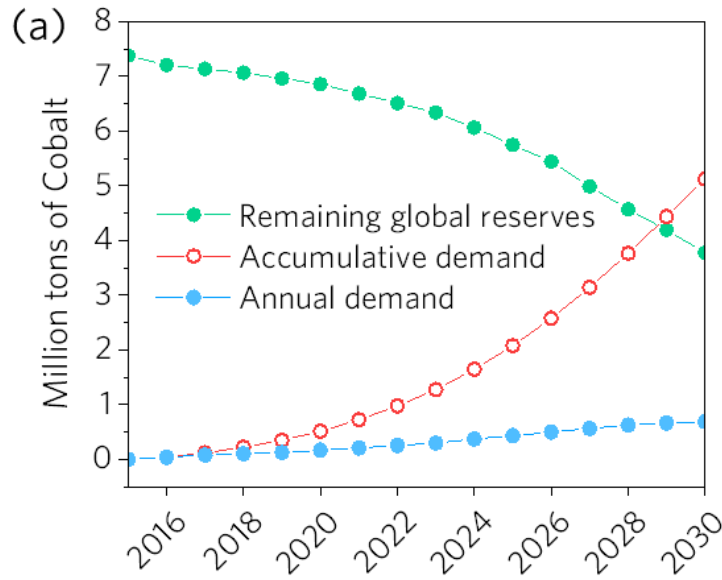
Anode Materials:

- Graphite, *Si*..

Lithium-ion batteries on the global market, forecast nameplate capacity, GWh



Global Projections of Critical Materials Reserves vs Current Demand



Li-Ion Battery Recycling Principles



DIRECT CATHODE RECYCLING

- Cathode Separation
- Binder Removal
- Relithiation
- Compositional Change



OTHER MATERIAL RECOVERY

- Electrolyte
- Graphite
- Foil



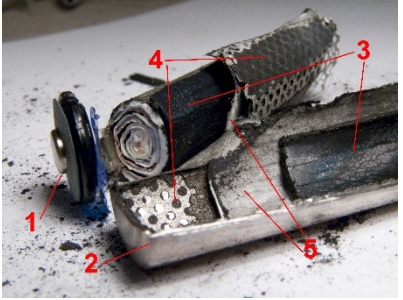
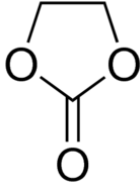
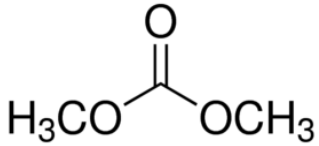
DESIGN FOR RECYCLING

- Cell Design
- Cell Rejuvenation

Direct Recycling



Conventional Recycling

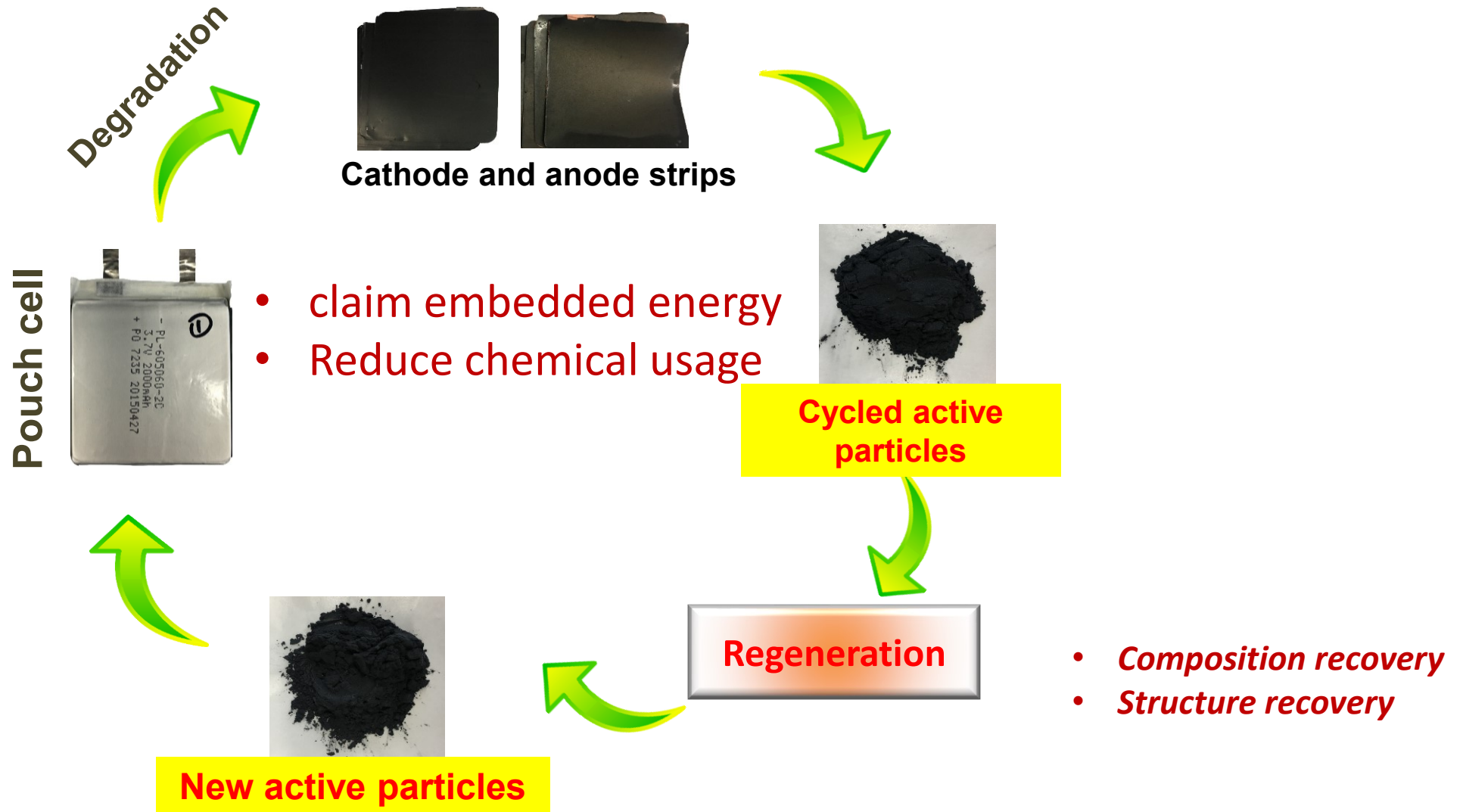


Ease of Recycling

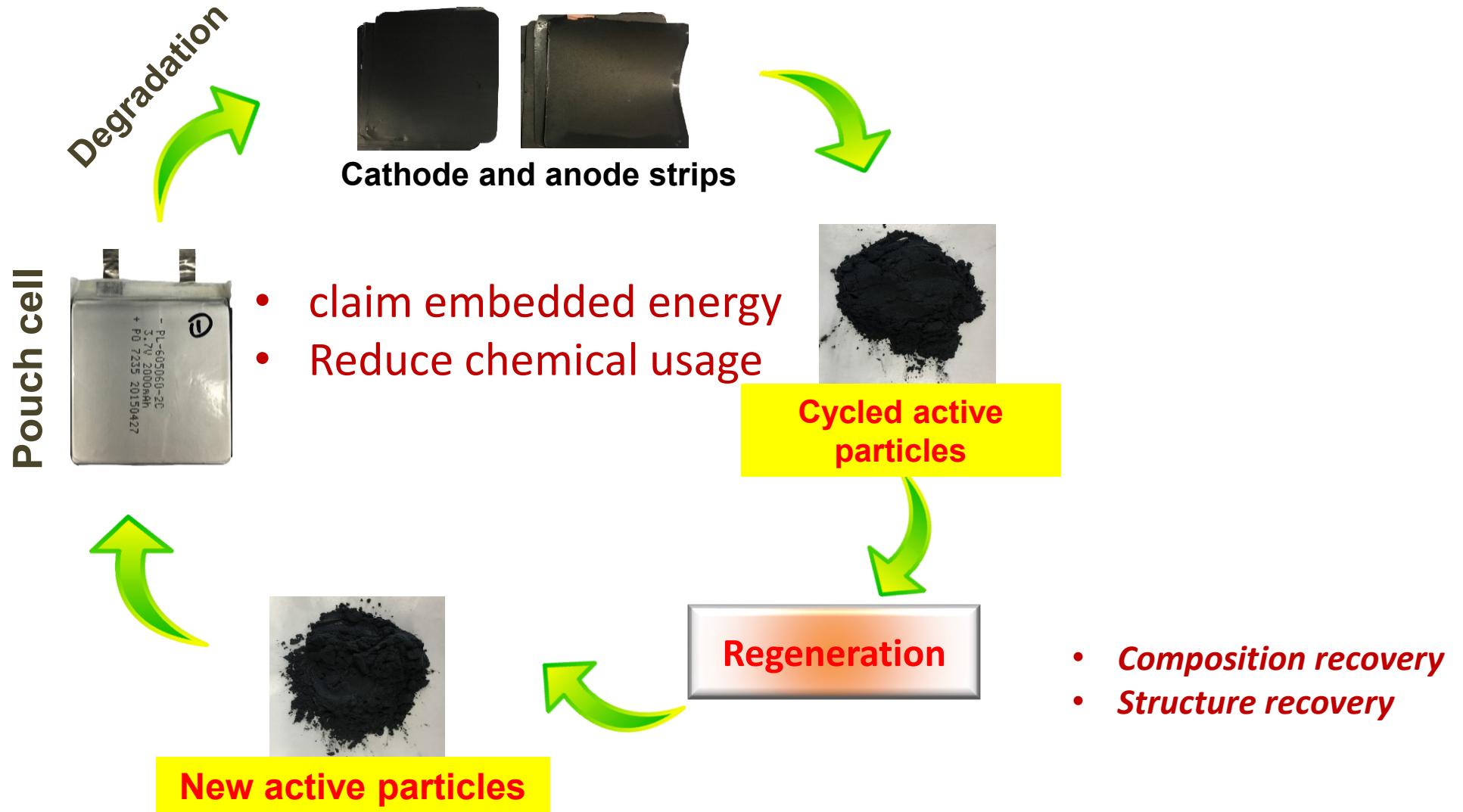


Better Labelling

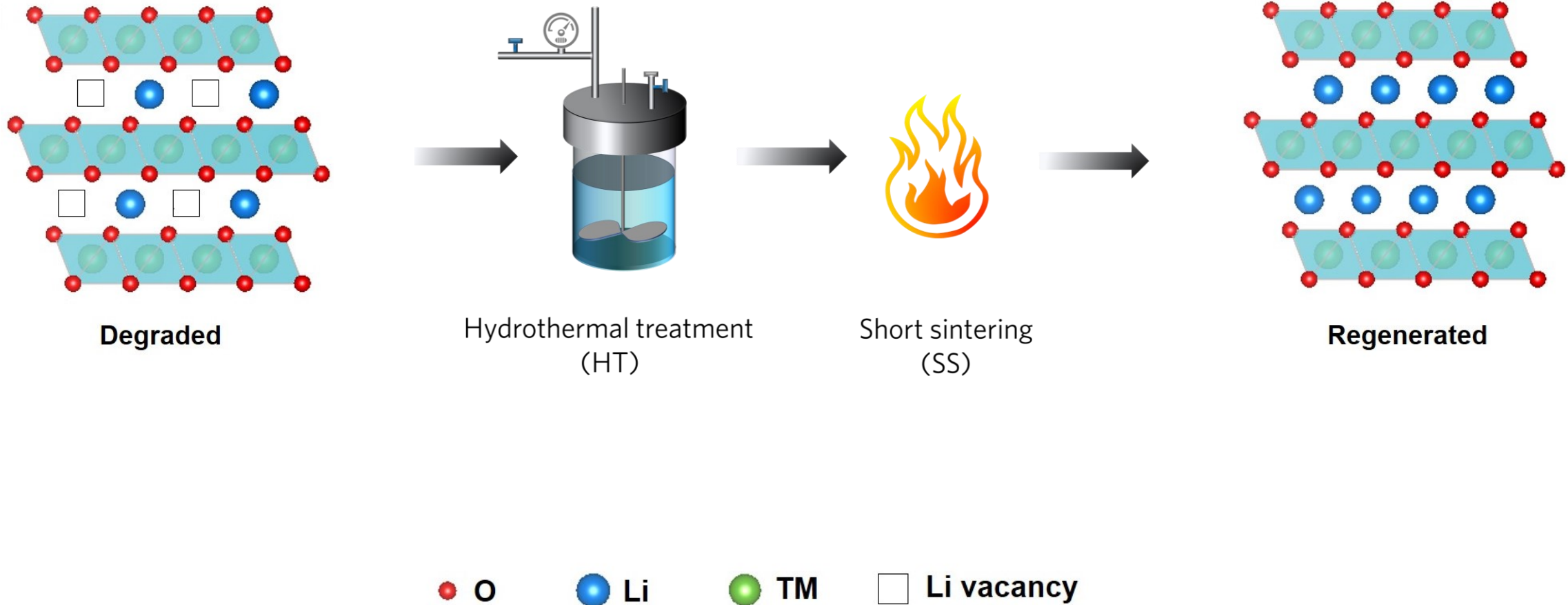
The “Direct” Recycling Process



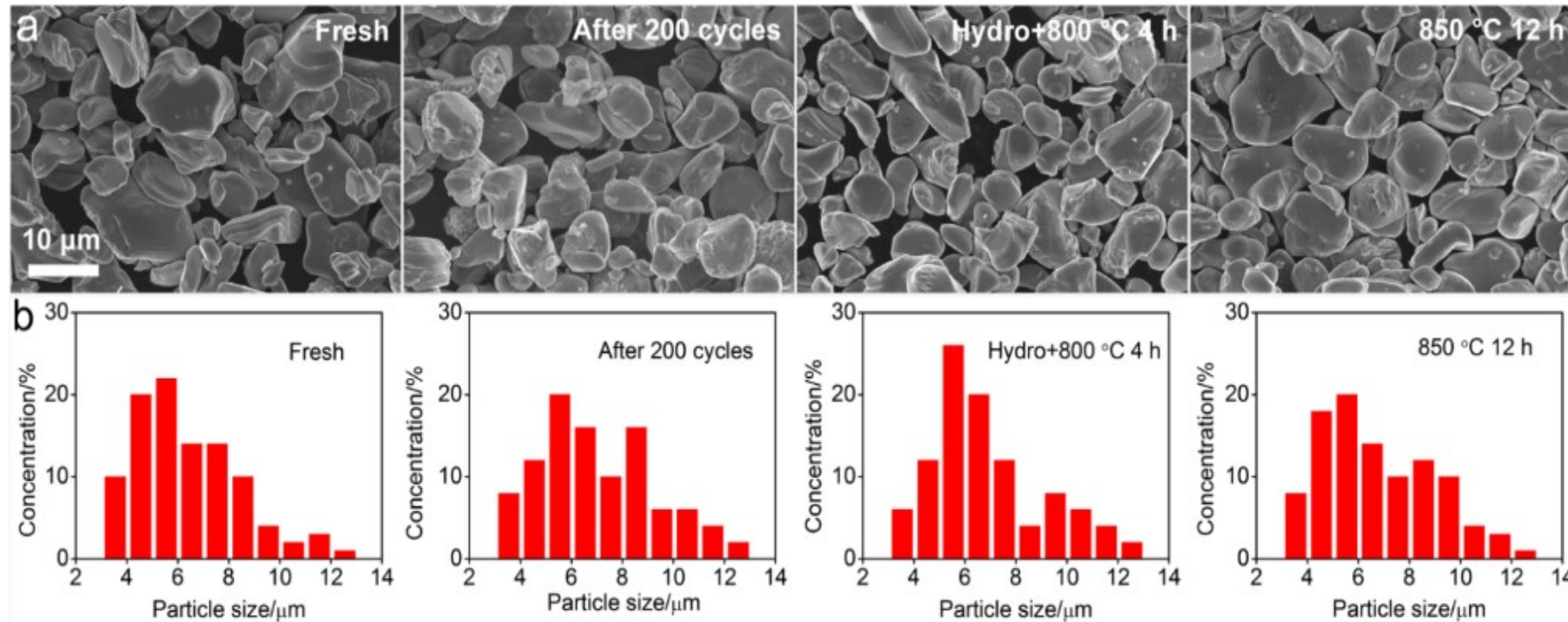
The “Direct” Recycling Process



The Direct Regeneration Process

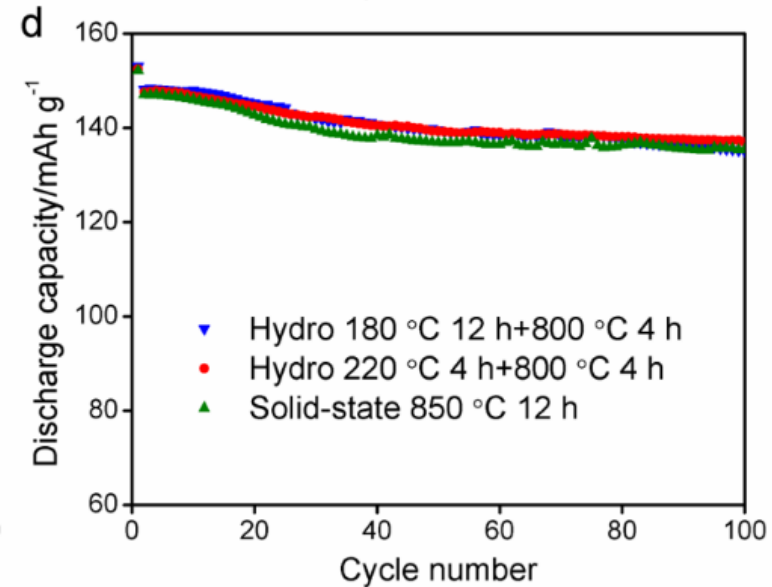
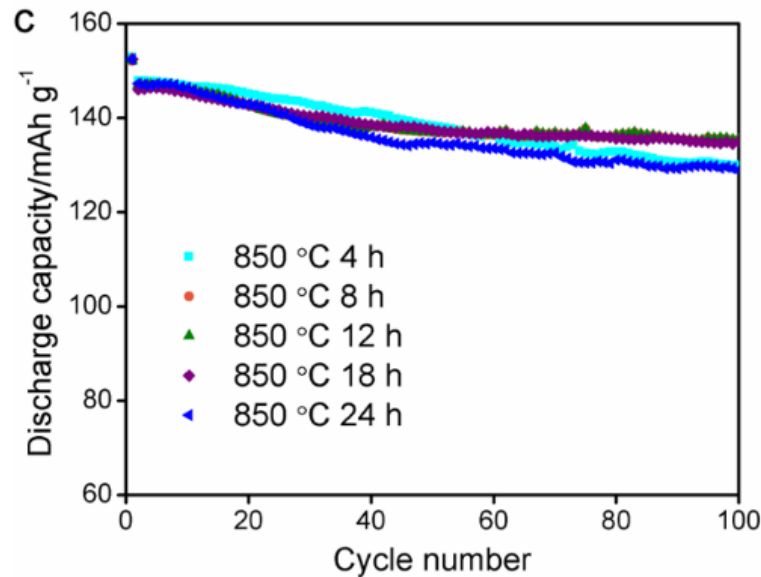
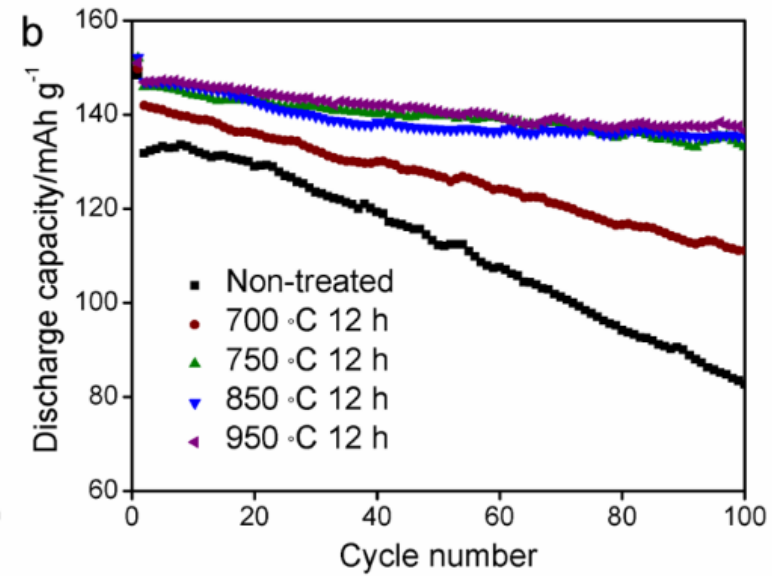
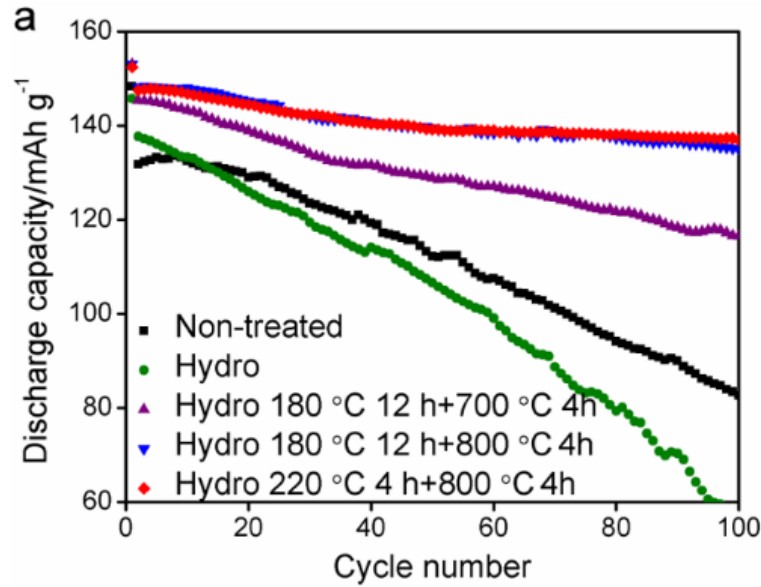


Morphological and Composition Change: the LCO Case

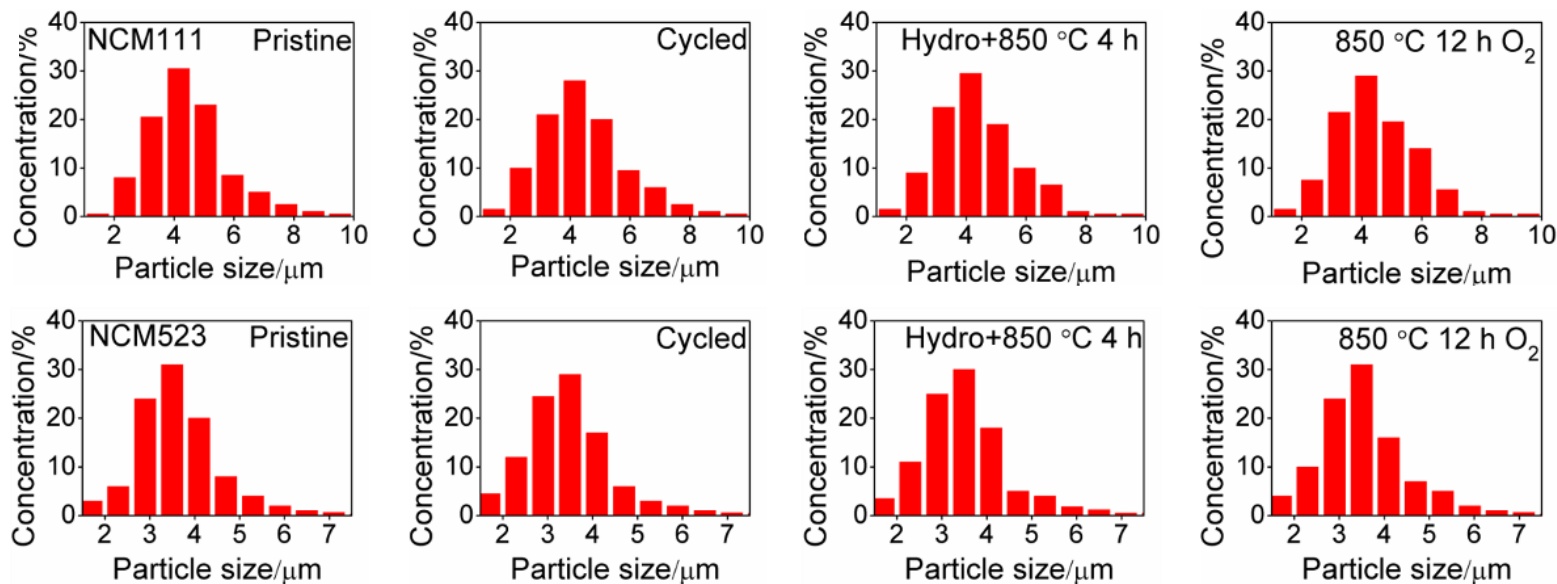
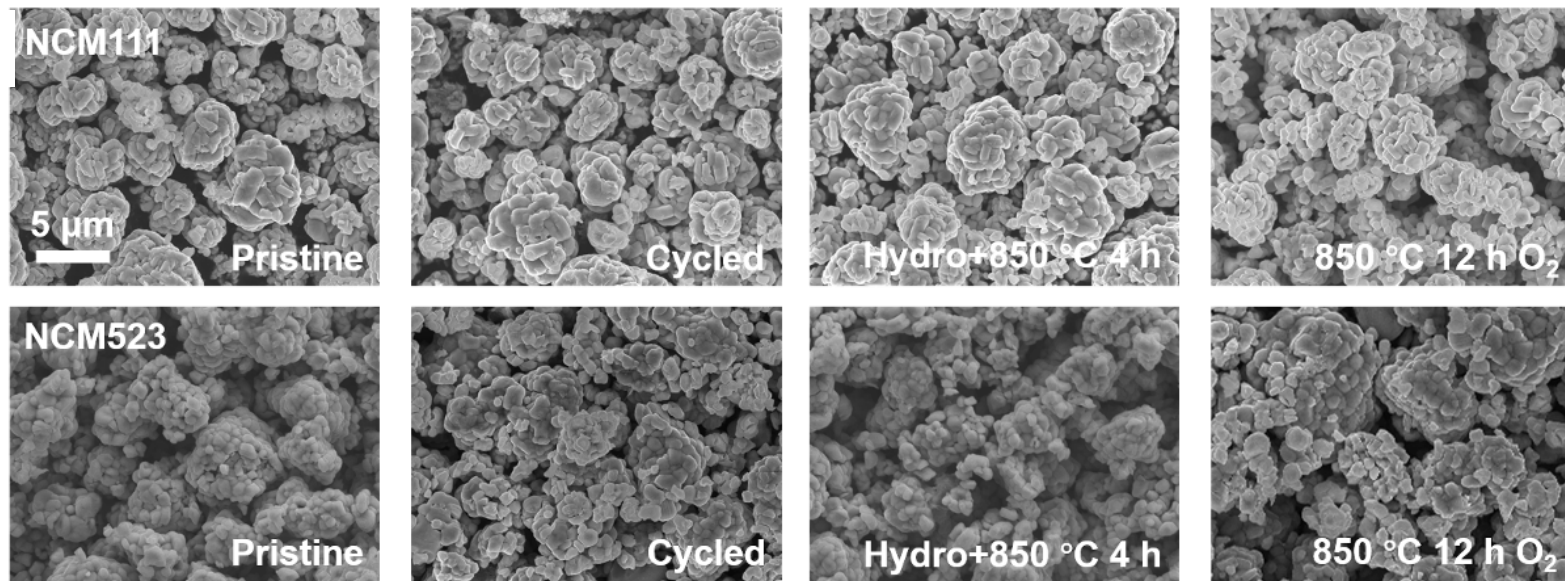


Hydrothermal	No annealing	700 °C 4 h	800 °C 4 h
Composition	$\text{Li}_{0.98}\text{CoO}_2$	$\text{Li}_{0.98}\text{CoO}_2$	$\text{Li}_{0.98}\text{CoO}_2$
Solid-state synthesis	750 °C 12 h	850 °C 12 h	950 °C 12 h
Composition	$\text{Li}_{0.99}\text{CoO}_2$	$\text{Li}_{0.98}\text{CoO}_2$	$\text{Li}_{0.96}\text{CoO}_2$

Exploring the LCO Regeneration Conditions



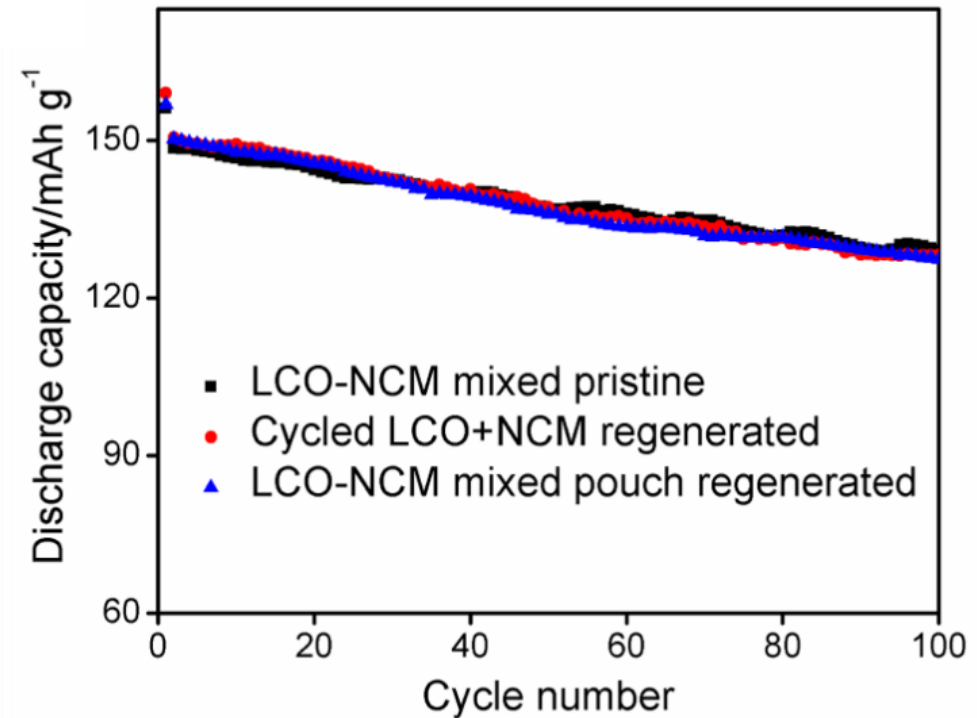
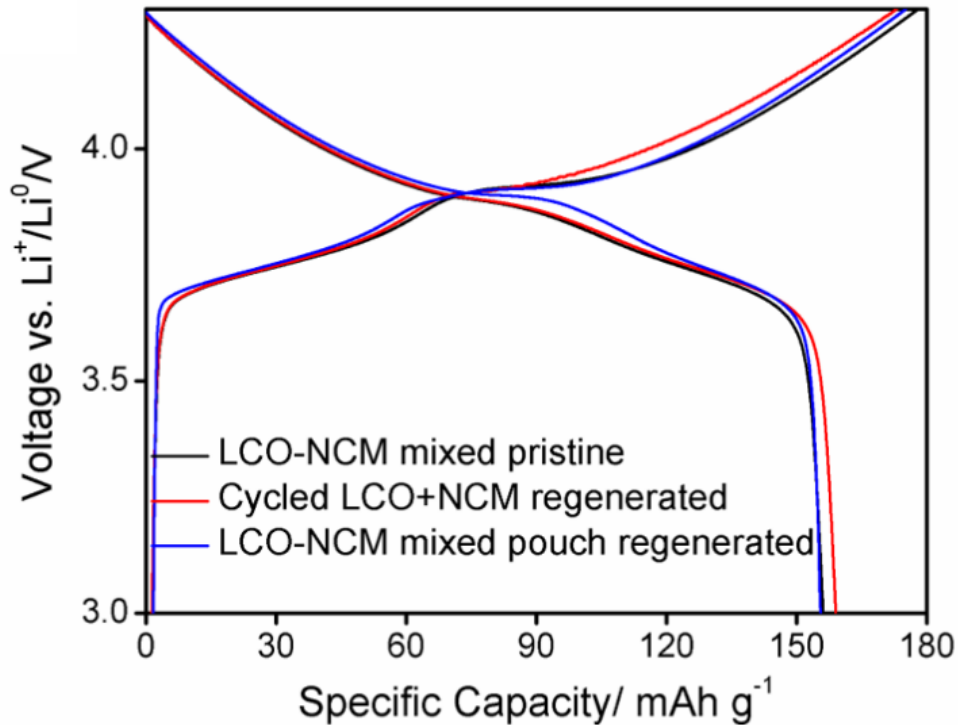
Recovery of Complicated NMC



Regeneration of $\text{LiNi}_x\text{Mn}_y\text{Co}_z\text{O}_2$ (NMC)

Sample	NCM111	NCM523
Pristine	$\text{Li}_{0.995}\text{Ni}_{0.331}\text{Co}_{0.341}\text{Mn}_{0.329}\text{O}_{2.012}$	$\text{Li}_{1.009}\text{Ni}_{0.492}\text{Co}_{0.209}\text{Mn}_{0.305}\text{O}_{2.015}$
Cycled	$\text{Li}_{0.786}\text{Ni}_{0.328}\text{Co}_{0.340}\text{Mn}_{0.325}\text{O}_{1.996}$	$\text{Li}_{0.788}\text{Ni}_{0.490}\text{Co}_{0.208}\text{Mn}_{0.302}\text{O}_{1.985}$
Hydro only	$\text{Li}_{1.012}\text{Ni}_{0.329}\text{Co}_{0.341}\text{Mn}_{0.326}\text{O}_{2.009}$	$\text{Li}_{1.006}\text{Ni}_{0.491}\text{Co}_{0.209}\text{Mn}_{0.304}\text{O}_{2.012}$
Hydro+850 °C 4 h	$\text{Li}_{1.019}\text{Ni}_{0.330}\text{Co}_{0.340}\text{Mn}_{0.327}\text{O}_{2.011}$	$\text{Li}_{1.021}\text{Ni}_{0.490}\text{Co}_{0.209}\text{Mn}_{0.303}\text{O}_{2.013}$
850 °C 12 h air	$\text{Li}_{1.016}\text{Ni}_{0.331}\text{Co}_{0.340}\text{Mn}_{0.325}\text{O}_{2.010}$	$\text{Li}_{1.017}\text{Ni}_{0.490}\text{Co}_{0.207}\text{Mn}_{0.304}\text{O}_{2.014}$
850 °C 12 h oxygen	$\text{Li}_{1.016}\text{Ni}_{0.331}\text{Co}_{0.341}\text{Mn}_{0.326}\text{O}_{2.012}$	$\text{Li}_{1.018}\text{Ni}_{0.491}\text{Co}_{0.208}\text{Mn}_{0.304}\text{O}_{2.013}$

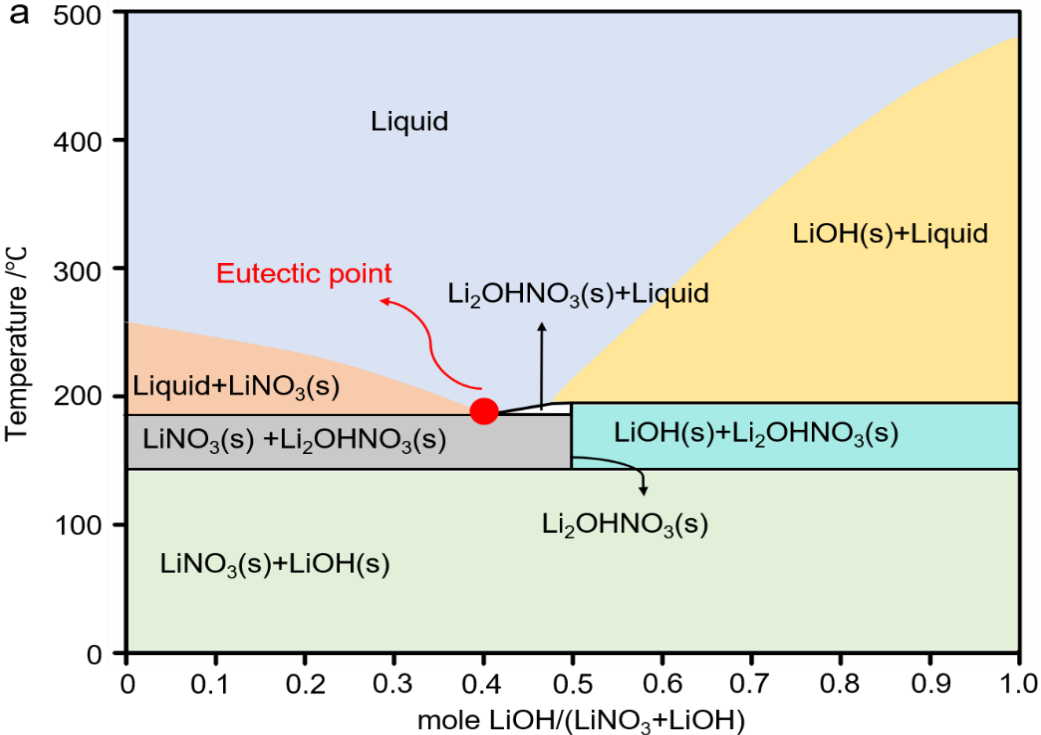
Regeneration of Mixed Cathodes



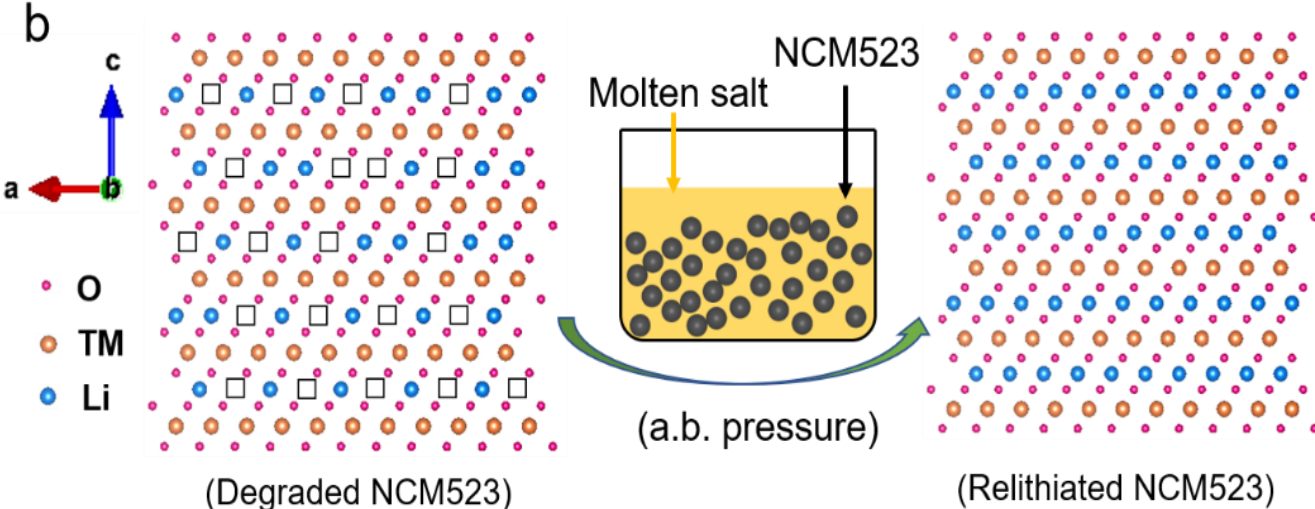
LCO-NCM mixed: materials mixed in the same cell

LCO+NCM mixed: cells with different materials and then mixed

Ambient Pressure Re-lithiation in Eutectic Mixtures

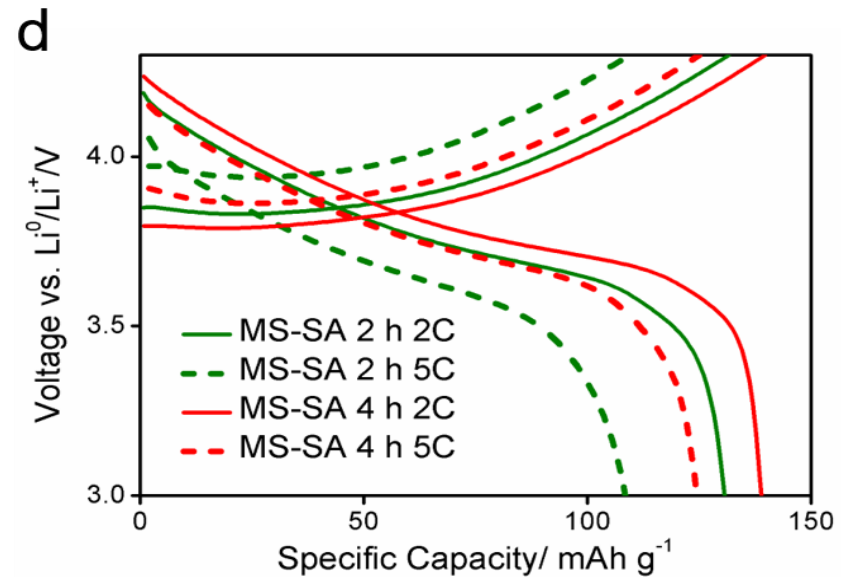
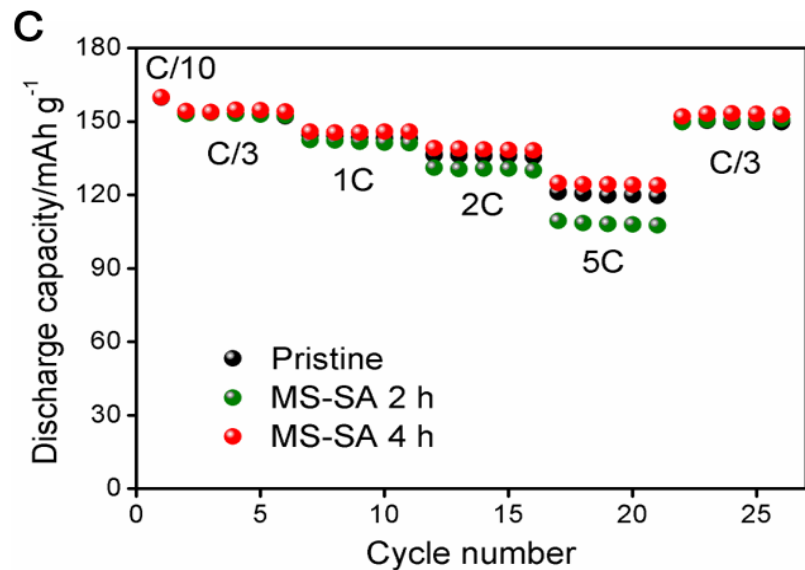
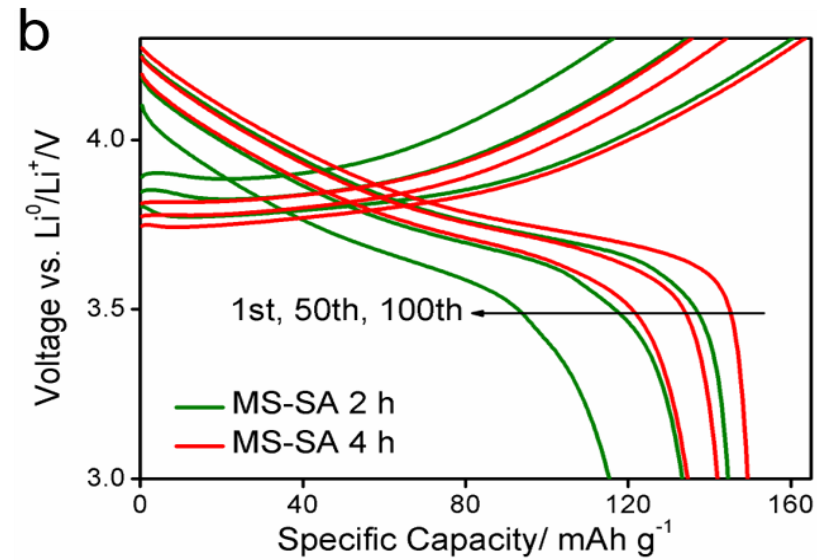
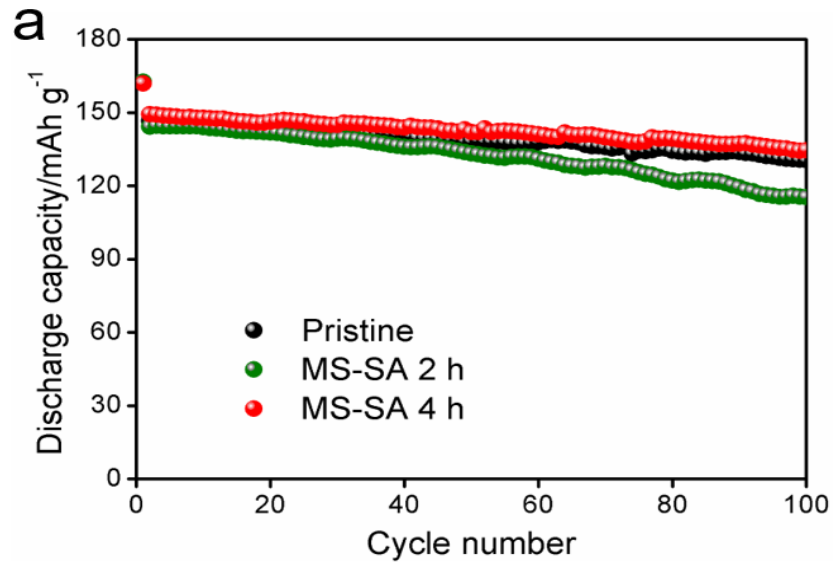


Relithiation of 50% degraded NCM

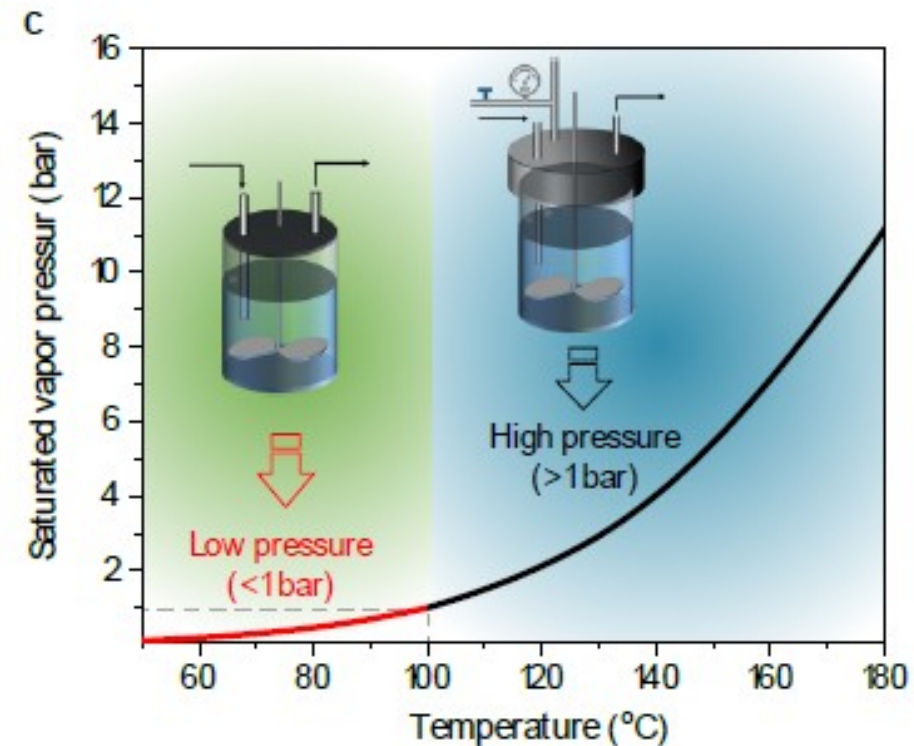
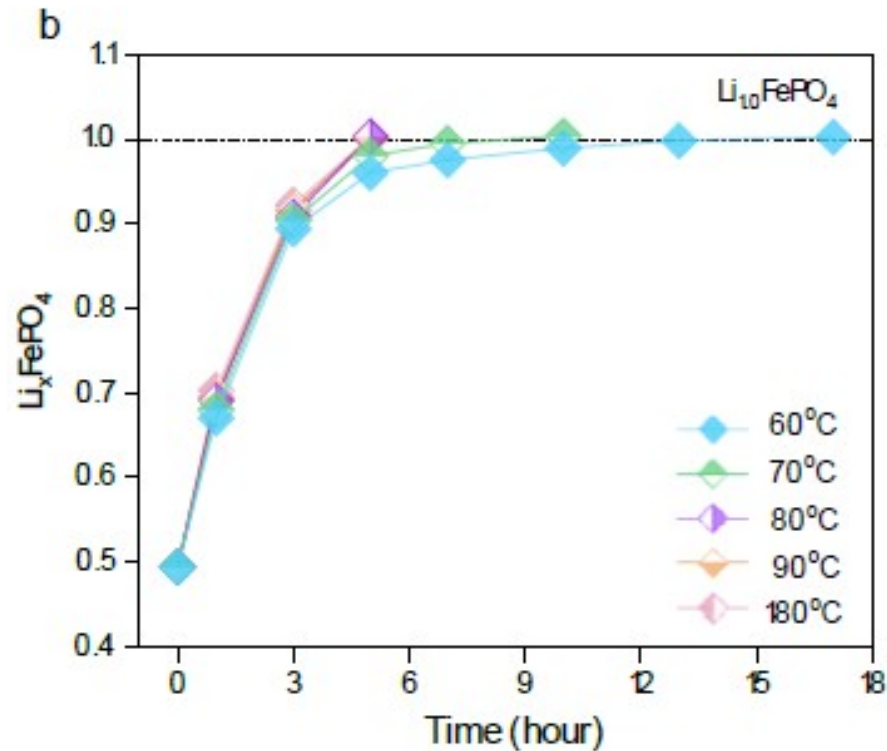
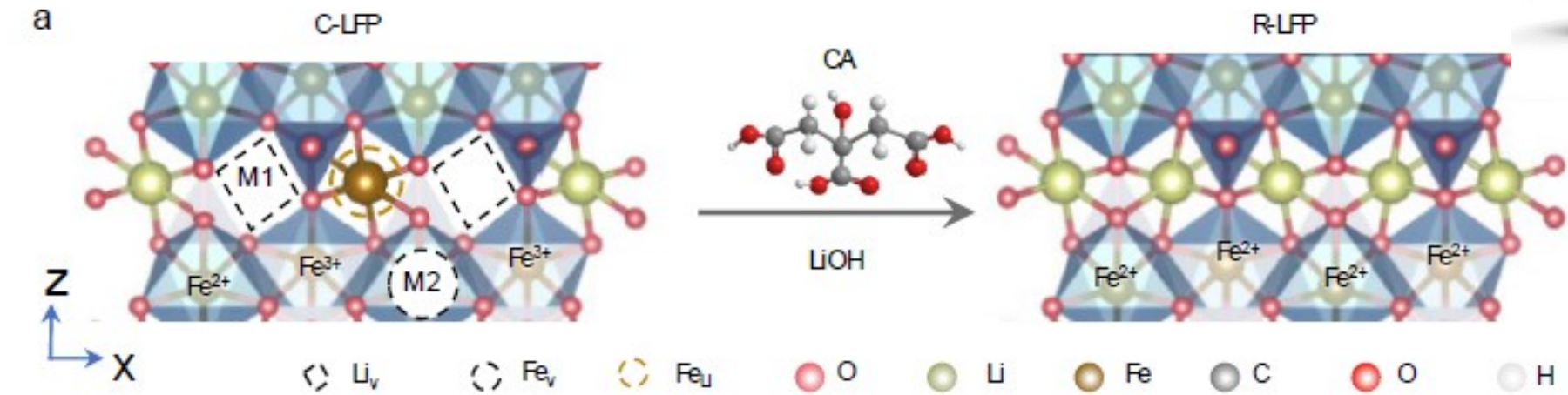


S. Yang, Z. Chen .et al, Adv. Energy. Mater, 2019, 9, 1900454.

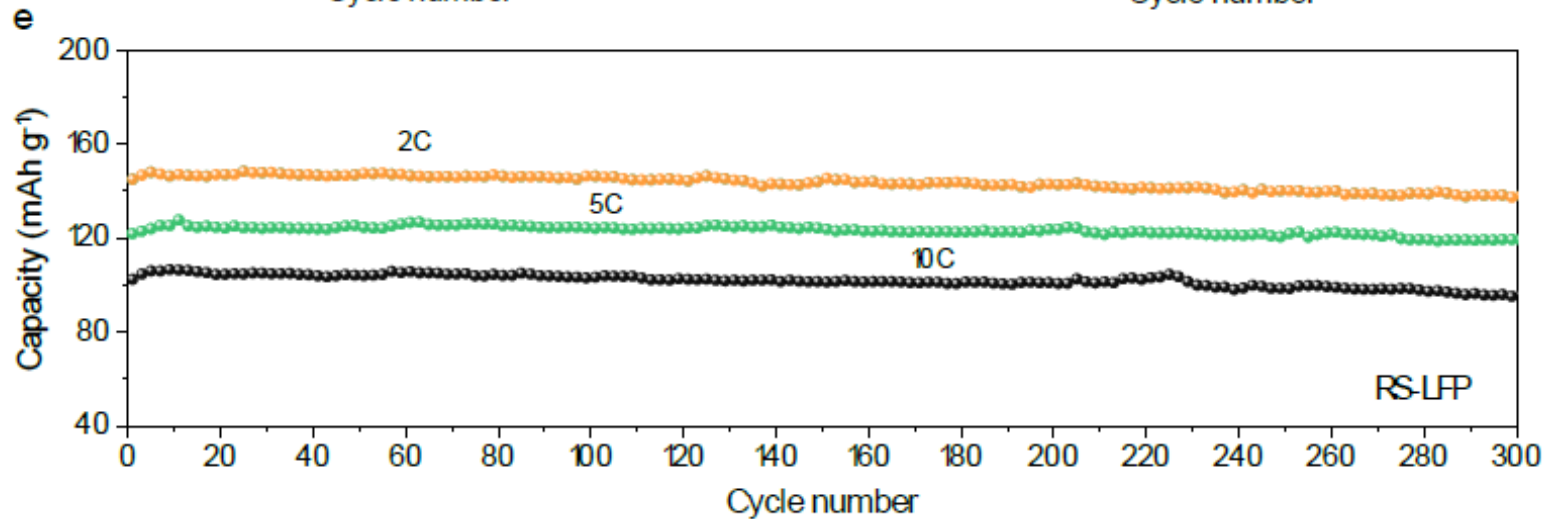
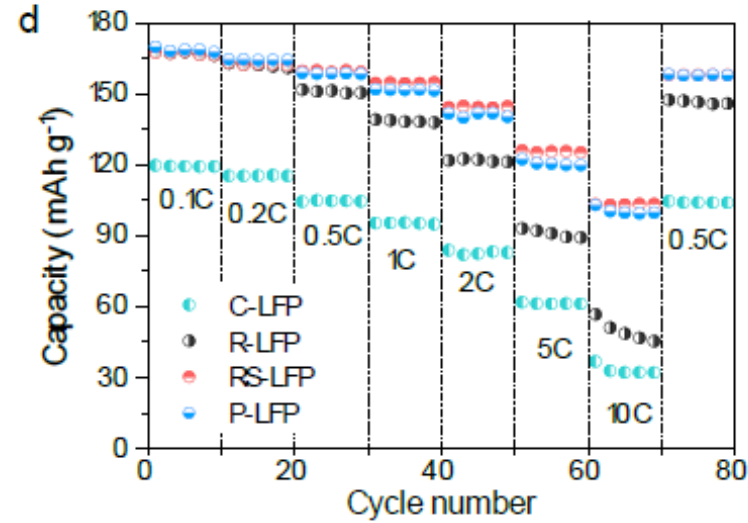
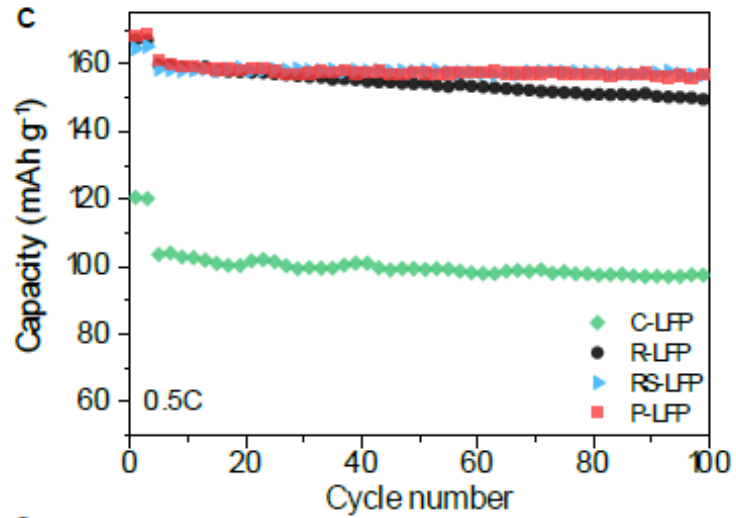
EC Data for Regeneration of NCM523



Direct Recycling of LiFePO_4

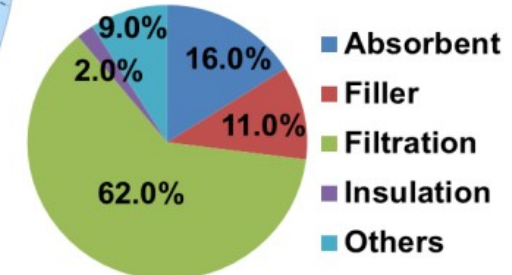
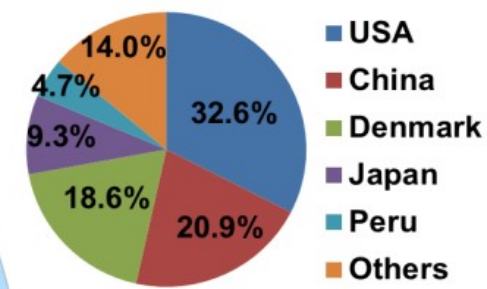
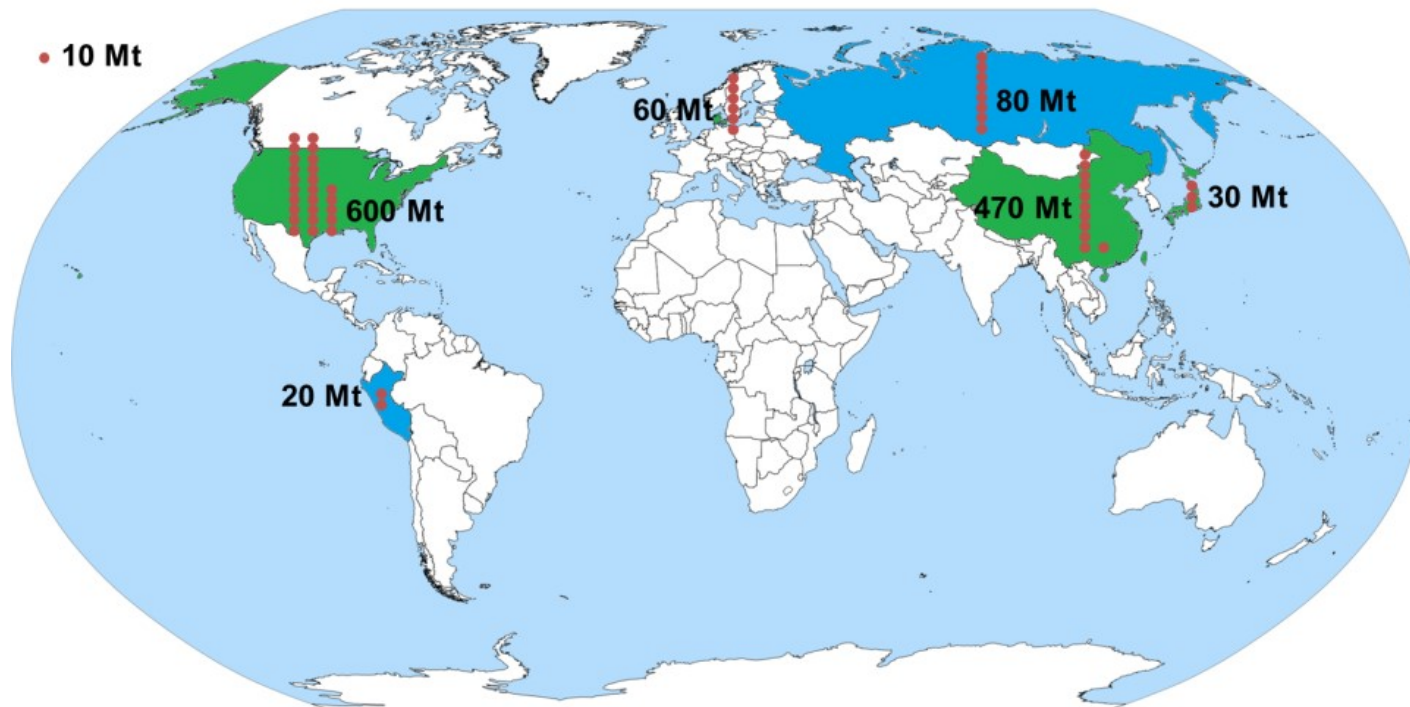
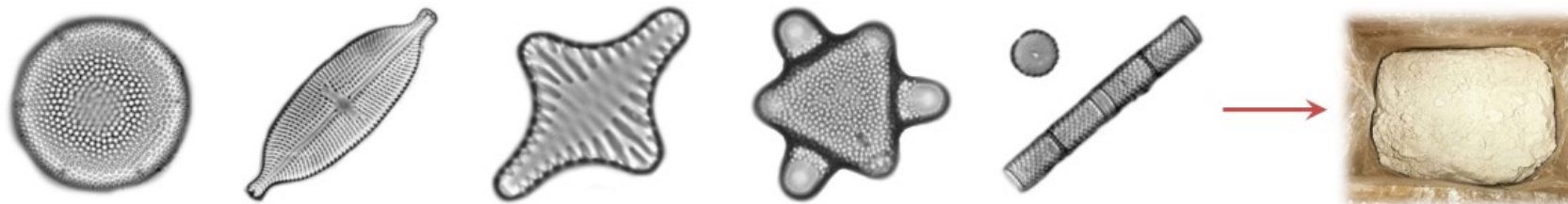


Performance of Reg. LiFePO₄



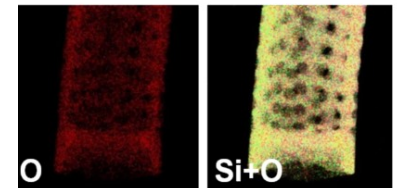
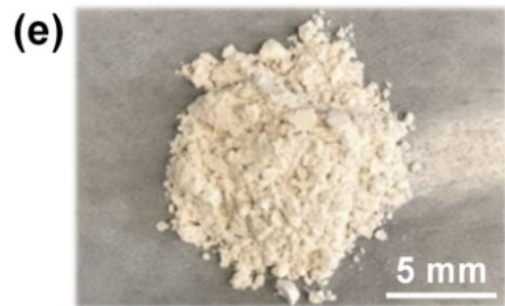
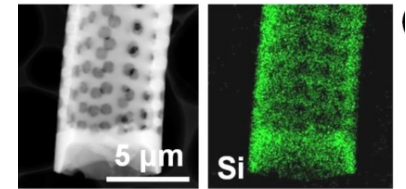
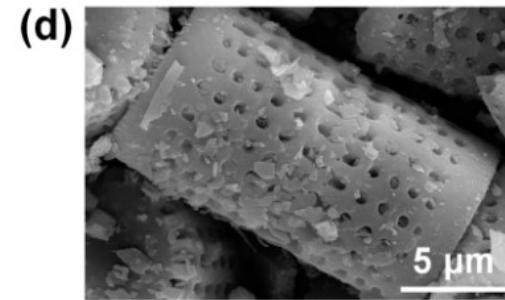
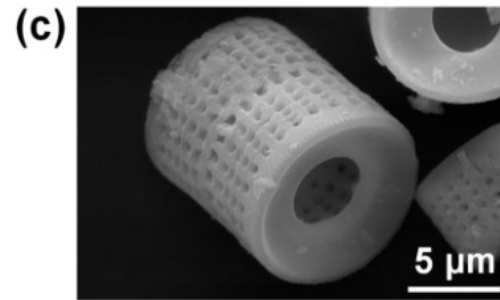
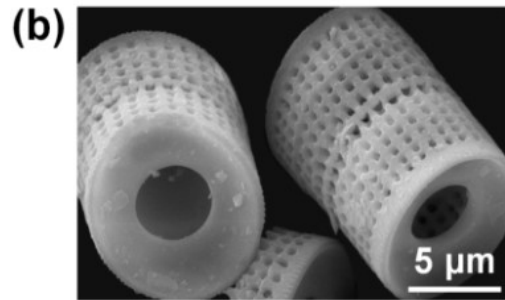
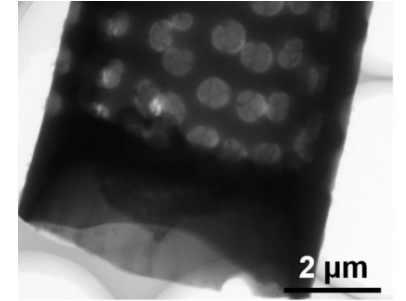
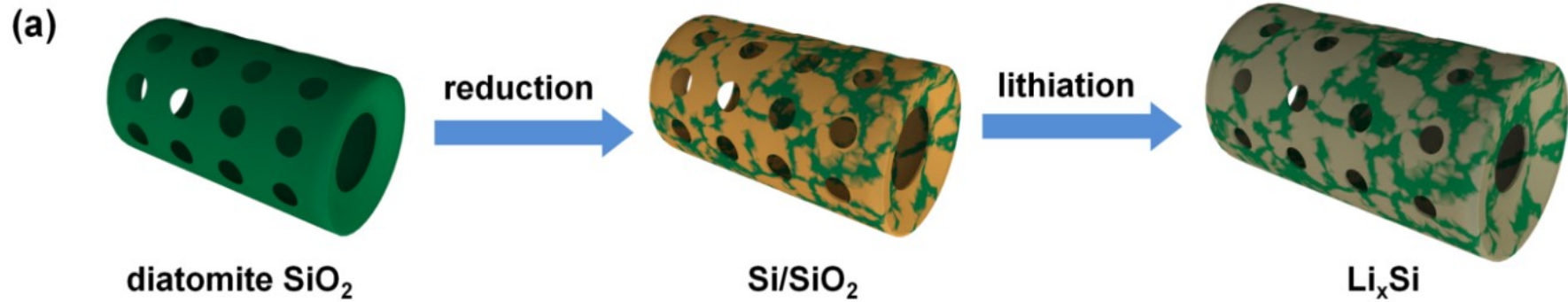
Z. Chen, et al, 2020, *Joule*,
doi.org/10.1016/j.joule.2020.10.008.

Diatomite: A Great Resource

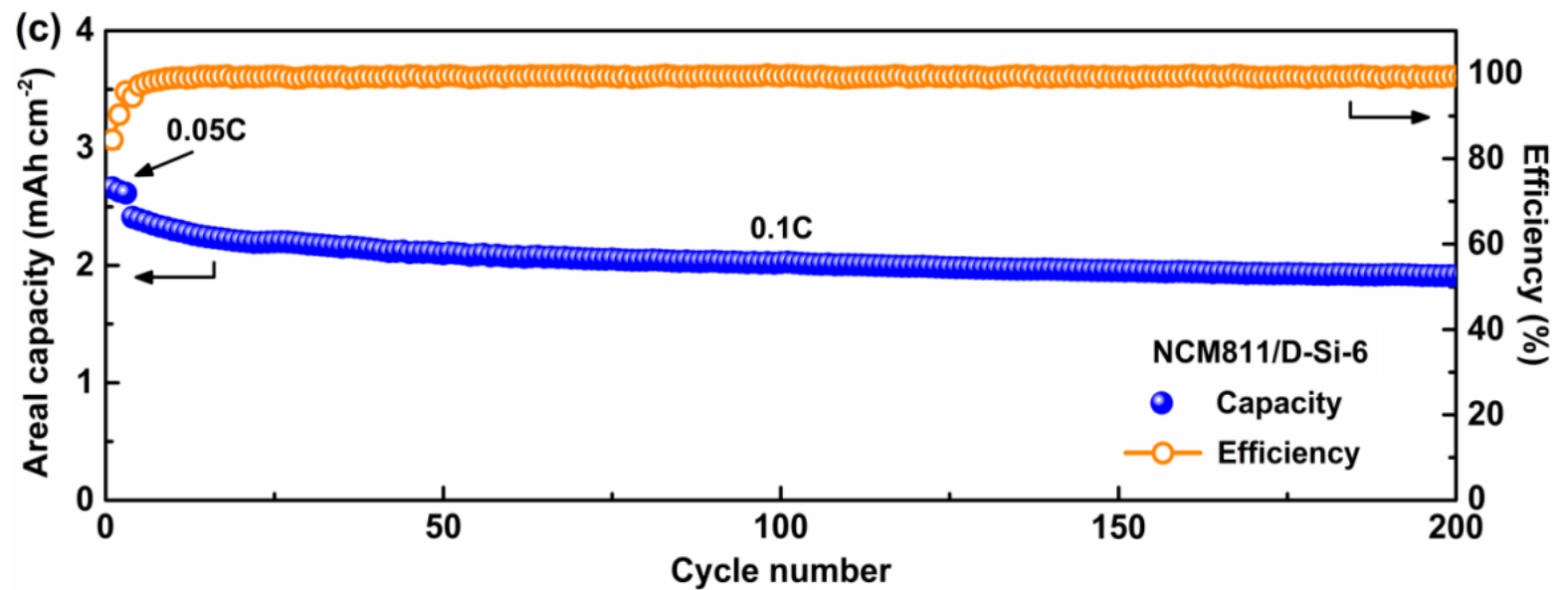
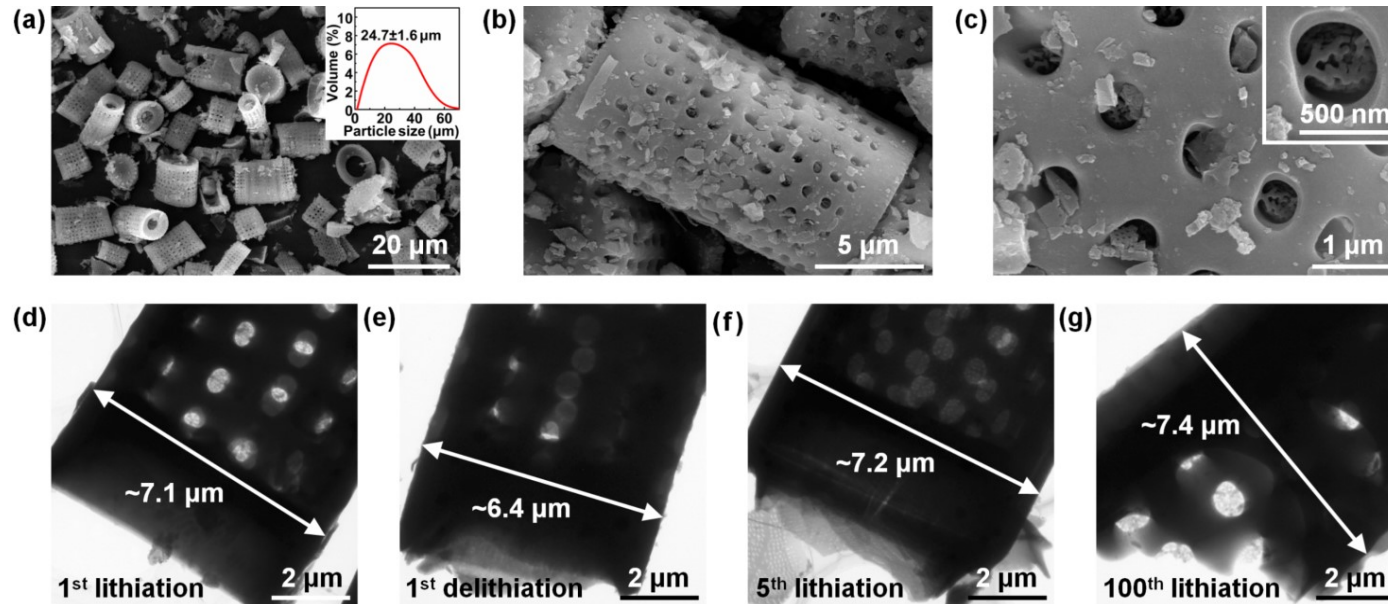


World proved reserve >2 billion ton, world output >3 million ton, low cost of 10~1000 \$/t

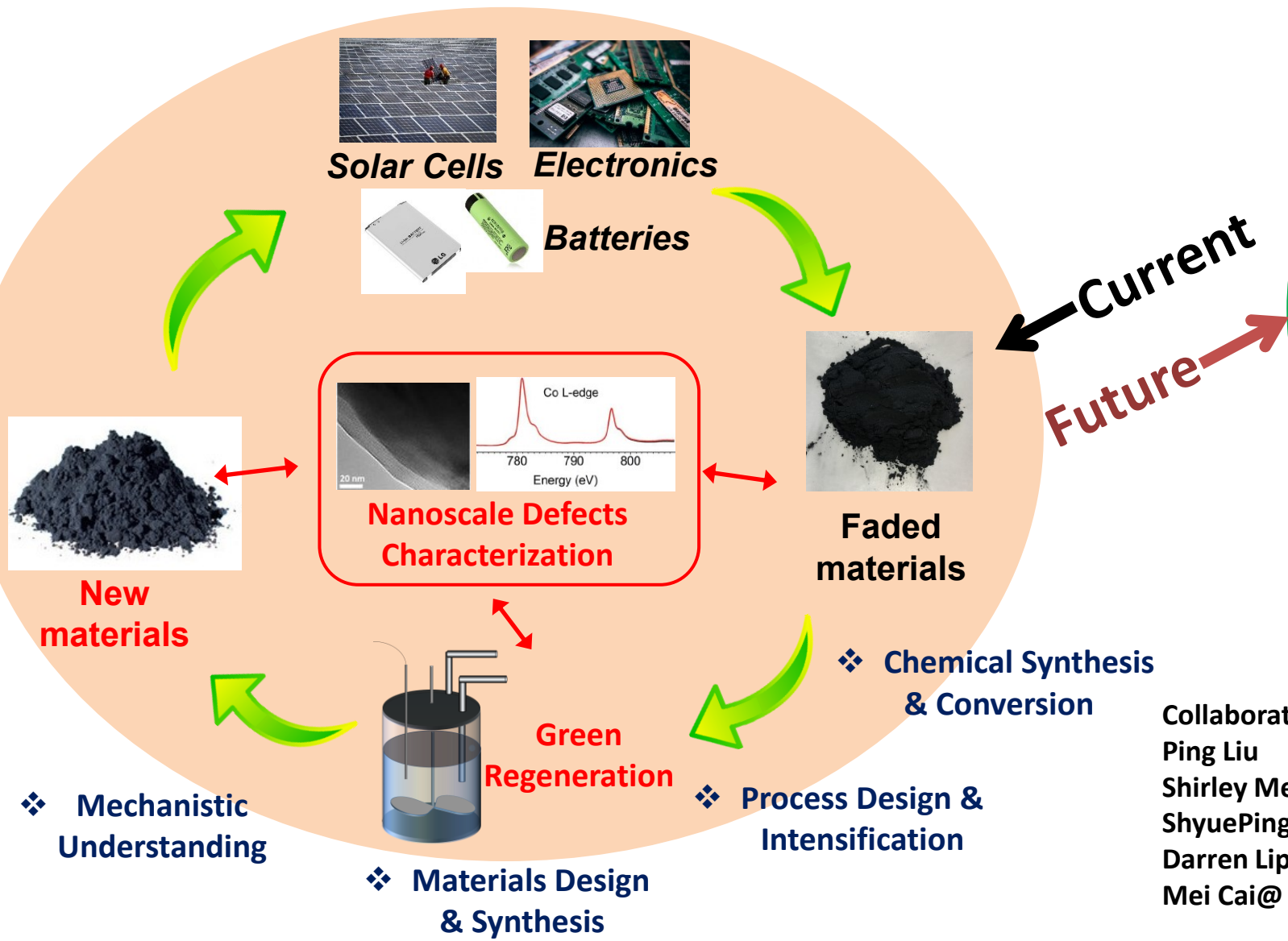
Diatomite-Derived Si Anode



Stable Cycling



Materials Science and Chemical Technology for Sustainable Energy



Current ←

→ **Future**



Collaborators:
 Ping Liu
 Shirley Meng
 ShyuePing Ong
 Darren Lipomi
 Mei Cai@ GM



<http://zhengchen.eng.ucsd.edu>

Funding: DOE, NSF, private companies.

Thank you!