Neutron diffraction and imaging on battery systems

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Li-ion battery: principle of operation
Market price $/kWh based on Li-ion technology

Initial price ~$4,000/kWh

Tesla Roadster ~$240/kWh

with Novel Chemistry & M

Basic Li-ion I


$1,200 $1,000 $800 $600 $400 $200 $0

Price [$/kWh]

Courtesy: Gleb Yushin
Projected production of Li-ion batteries in Europe

Over 300 GWh/a Li-Ion Battery Cell Production Capacity Announced in Europe

- **Salzgitter, 2024**
  - 16 GWh, later 24 GWh
- **Erfurt, 2022**
  - 14 GWh, later 100 GWh
- **Sunderland, 2010**
  - 2.5 GWh
- **Willstät, 2020**
  - 1 GWh
- **France, 2022**
  - Capacity unknown
- **Germany, 2023**
  - 20 GWh, later 24 GWh
- **Germany, 202X**
  - 4 GWh, later 8 GWh
- **Mo i Rana, 2023**
  - Ramp up to 32 GWh
- **Skelleftea, 2021**
  - 8 GWh, later 32 GWh
- **Bitterfeld, 2022**
  - 10 GWh
- **Wroclaw, 2018**
  - 6 GWh, later 70 GWh
- **Komarom, 2020**
  - 7.5 GWh
- **Göd, 2018**
  - 3 GWh, later 15 GWh
- **Europe, 202X**
  - Capacity unknown
- **Europe, 202X**
  - Capacity unknown

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Moore's law does not hold true for battery development

- 120 mAh/annum average increase rate over almost 25 years
- ~ 3.5%/year relative increase
- Since 2012 the capacity increase is achieved by voltage increase and introduction of Si to graphite anodes

Not steady increase, but a step in performance of Li-ion batteries is required... pure material problem
Materials for battery applications

**Anode materials**
- \( \text{Li}_4\text{Ti}_5\text{O}_{12} \)
- \( \text{Li}_2\text{TiO}_3 \)

**Cathode materials**
- \( \text{Li}_{1-x}\text{CoPO}_4 \)
- \( \text{Li}_x\text{MoO}_2 \)

**Lithium electrolytes**
- \( \text{Li}_{1.3}\text{Al}_{0.3}\text{Ti}_{1.7}(\text{PO}_4)_3 \)
- \( \text{Li}_{10}\text{GeP}_2\text{S}_{12} \)
Different mechanisms of Li-ion battery degradation
Neutron-based experimental techniques with proven relevance\impact in battery research

**Neutron diffraction**: detail of crystal structure, localisation and quantification of lithium; microstructural studies; phase analysis.

**Neutron imaging**: lithium distribution, gas formation, electrolyte dynamics;

**Small-angle neutron scattering**: in-situ materials morphology and fracturing upon cell fatigue;

**Quasielastic neutron scattering**: in-situ structure and mobility of electrolytes in Li-ion batteries;

**Reflectometry**: studies of solid-electrolyte interphase; studies of lithiation in amorphous silicon; solid-liqued interfaces;

**Neutron depth profiling**: nanometer sensitive probe of lithium concentration in electrode materials;

**Positron spectroscopy**: charge- and fatigue-induced defect formation;

**Neutron and Prompt gamma activation analysis**: non-destructive and simultaneous elemental/isotope analysis;
Gas evolution in pouch cells studied by neutron radiography

4D imaging on lithium-batteries

Pristine/0 s
Partly dc/1500 s
-225.71 mAh
Partly dc/3900 s
-580.55 mAh

R.F. Ziesche et al., Nature Communications 11 (2020) 777
Simultaneous neutron radiography and diffraction data collection on 18650-type cell cycled up-side-down

Fresh cell

Fatigued cell

https://www.youtube.com/watch?v=ICPzHO_1nQ8
Why graphite?

Neutron, $\lambda=1.5482$ Å

High-energy synchrotron, $\lambda=0.20708$ Å
Simultaneous neutron radiography and diffraction data collection on 18650-type cell cycled up-side-down

https://www.youtube.com/watch?v=ICPzHO_1nQ8
Spatially-resolved neutron diffraction and current distribution in Li-ion batteries
Selected diffraction patterns
Lithium distribution in the middle of 18650-type cell
Spatially-resolved TOF neutron diffraction

Fresh
120 cycles
200 cycles
400 cycles

D. Petz et al., J. Power Sources 448 (2020) 227466
Spatially-resolved diffraction using conical slits

Gauge volume
Neutron diffraction

Gauge volume
Diffraction tomography

1.0 mm

Principle of diffraction tomography
Lithium distribution in the graphite anode of 18650-type lithium ion battery
Summary

- Perspectives for neutron powder diffraction and spatially-resolved diffraction
  
  *More flux, better resolution*

- Neutron diffraction tomography
  
  *Low-divergent\parallel monochromatic neutron beam with submillimeter focusing*

- Neutron imaging in battery research
  
  *More flux, better resolution, higher neutron energy*
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Neutron CT reconstruction from 18650-type LCO|C cell

ANTARES@FRMII

3.0 V

4.2 V

X-ray CT

poly

mono

ca. 30 µm
Neutron CT reconstruction from 18650-type LCO|C cell

SOC
- SOC 0
- SOC 100
- difference

Fatigue
- SOC 100, fresh
- SOC 100, fatigued
- difference
Evolution of the neutron diffraction signal (background subtracted) upon cooling of LP30 electrolyte filled in a thin-wall vanadium container.

Chosen electrolyte: LP30, EC+DMC+1M LiPF₆; Melting temperature: ca.250 K
Distribution of lithium and electrolyte concentration in fresh and aged 18650-type cells

**Lithium concentration** $x$ in $\text{Li}_x\text{C}_6$

- Fresh
- 600 cycles

**Electrolyte concentration** $m$

- Fresh
- 600 cycles