Estimating the Graphite Electrode Phase Evolution from Electrode Potential Measurements

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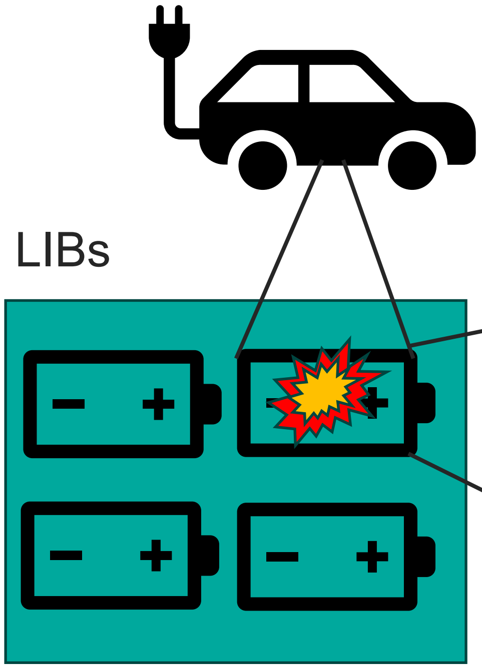
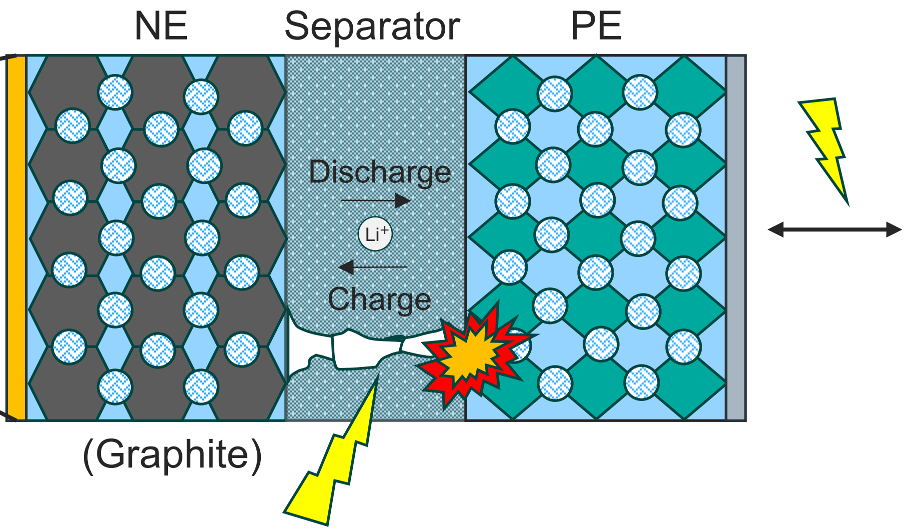
Materials & Design, 266:116218, 2026.

Introduction

Lithium ion cells consist of
- Negative electrode (NE)
- Separator
- Positive electrode (PE)

If charging current is too high, **lithium plating** may happen





Lithium ion batteries

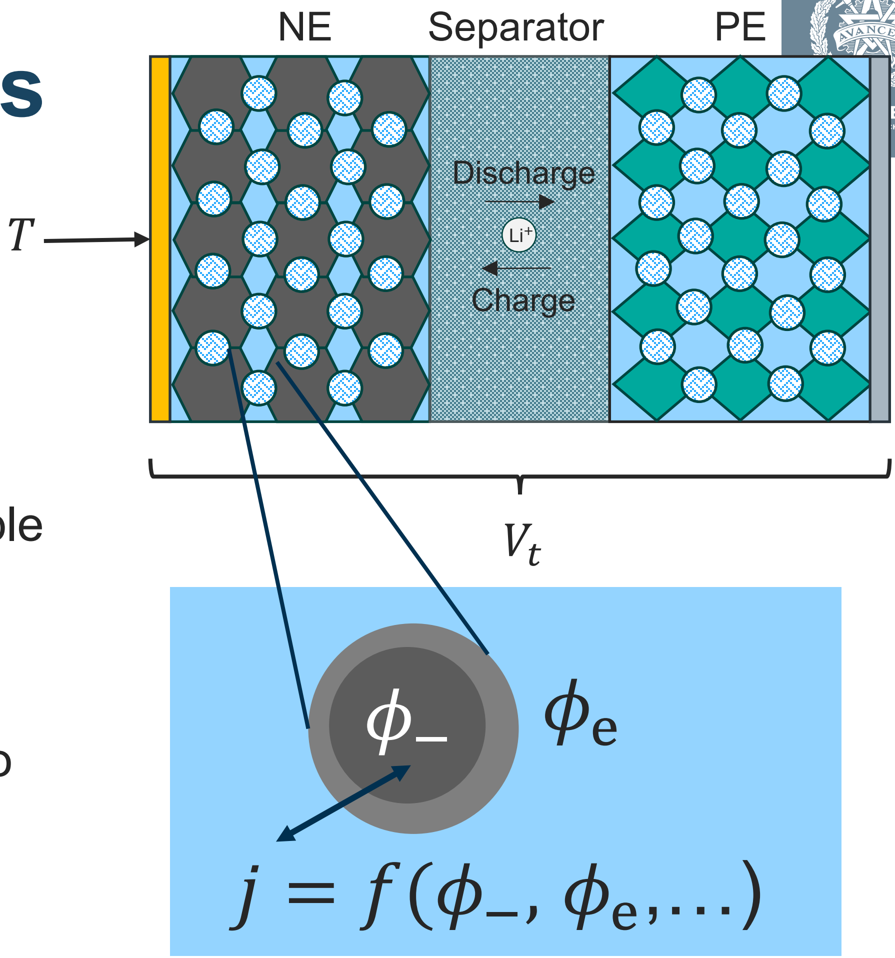
LIBs are **chemical systems**

- Electrode reactions are driven by electrode-electrolyte potential differences

Only external signals are measurable

- Surface temperature
- Voltage

Internal states must be estimated to achieve safe and optimal control



Phase evolution of graphite

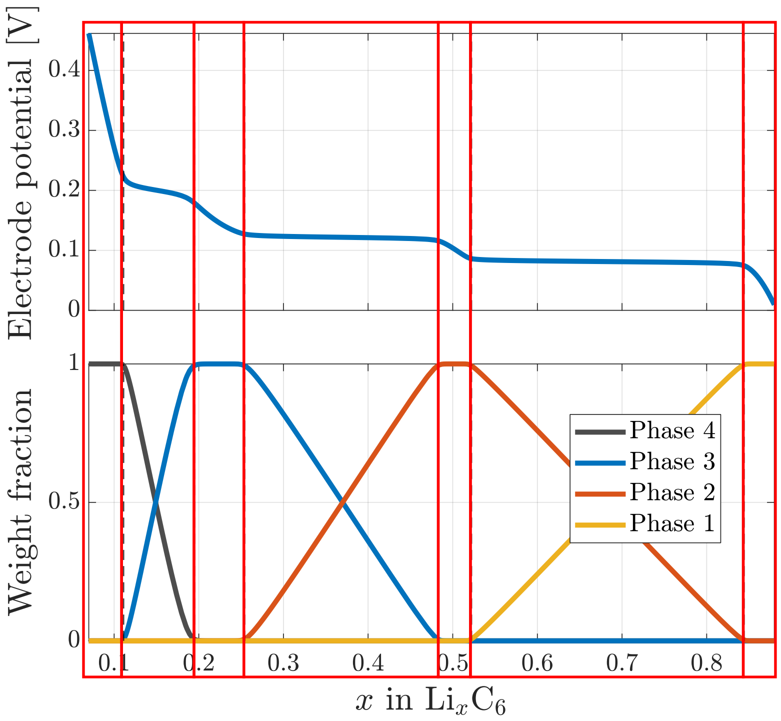
NE transitions through different molecular phases during lithiation

Different phases have different properties

- Potential
- Diffusivity

In final phase lithium plating is more likely

→ Keeping track of phase content is of interest

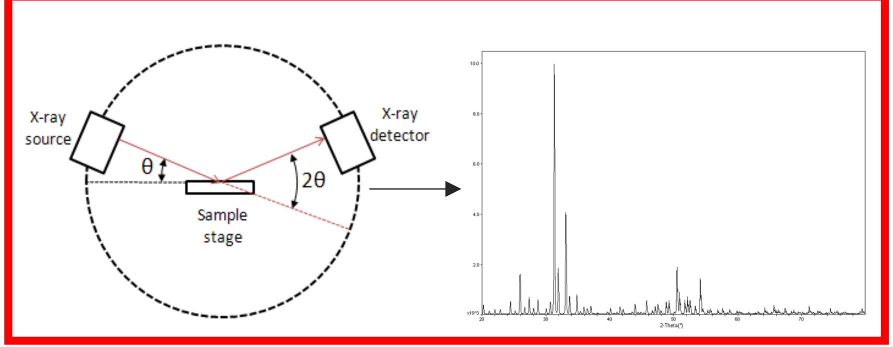


Y. Huang, Q. Zhu, T. Wik, D. P. Finegan, Y. Li, C. Zou (2026). BMINN: Learning chemical potentials and parameters from voltage data for multi-phase battery modeling. *Energy Storage Materials*, 86:104997.

Measuring the phase content

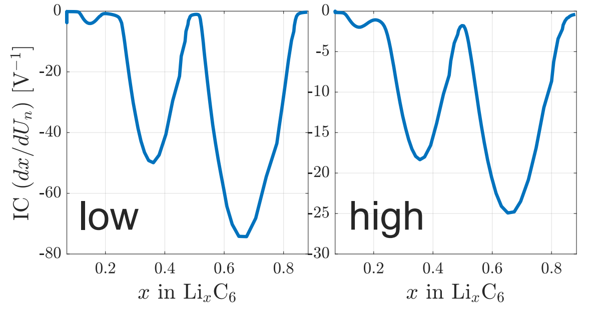
State of the art

- X-ray diffraction (XRD)
- Incremental Capacity Analysis (ICA) dQ/dV



XRD

- Expensive, not applicable in commercial devices
- Commercial cells can typically not be used



ICA

- Only accurate for low currents

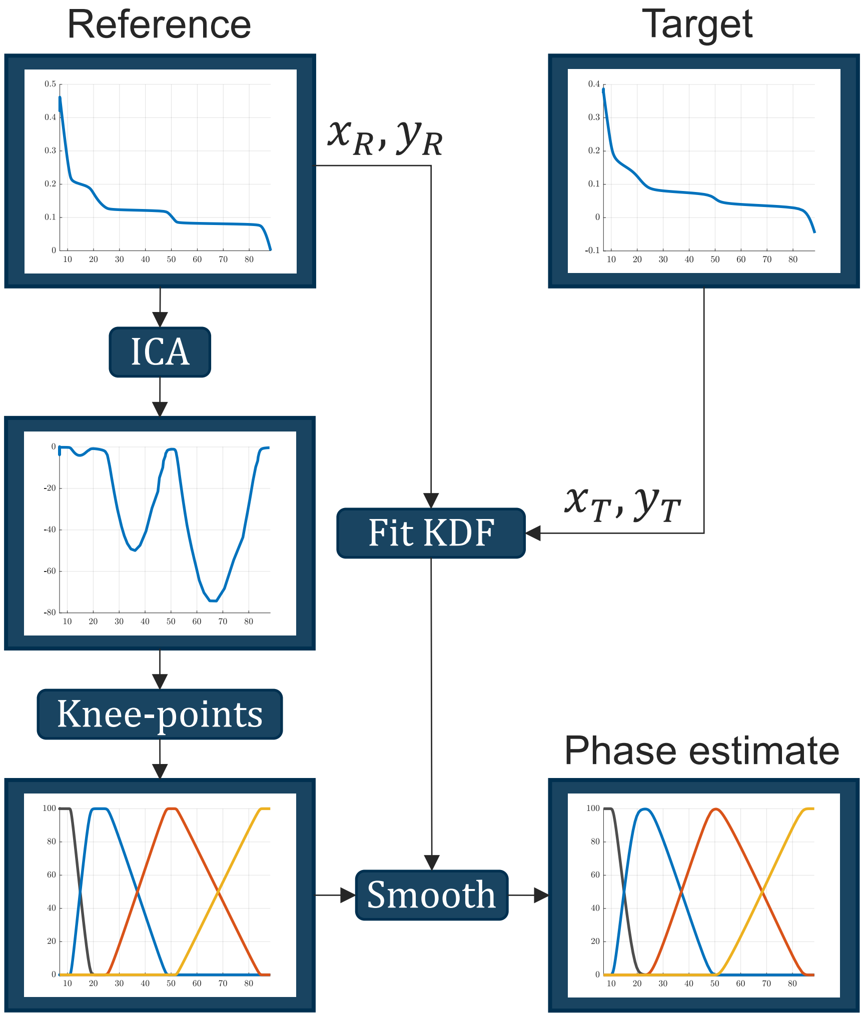
 Powder X-ray Diffraction. (2023, August 29). https://chem.libretexts.org/@go/page/314.

Estimation method

1. Measure data
- Reference: low current
- Target: any higher current

2. Reference phase estimation
- Calculate ICA
- Find knee-points
- Piecewise linear approximation

3. Kernel smoothing
- Fit kernel density function (KDF)
- Apply fitted KDF to reference phase estimate



Estimation at low currents

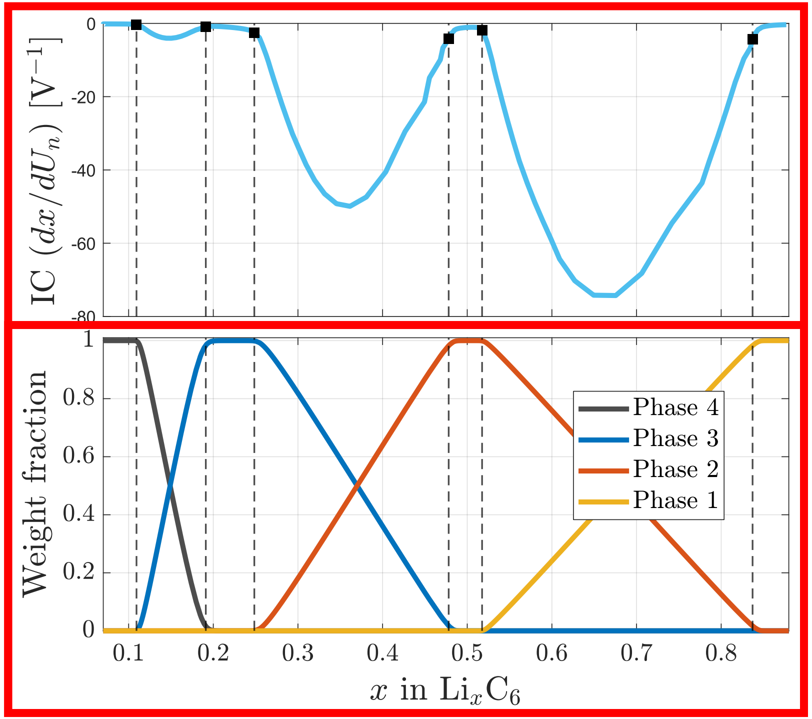
Low current:

- Linear phase changes
- Two phases active

→ Knee points determine phase evolution

Knee points in ICA approximate knee points in phases

→ Can be used for piecewise linear approximation



D. W. Bacon, & D. G. Watts (1971). Estimating the transition between two intersecting straight lines. *Biometrika*, 58(3):525–534.

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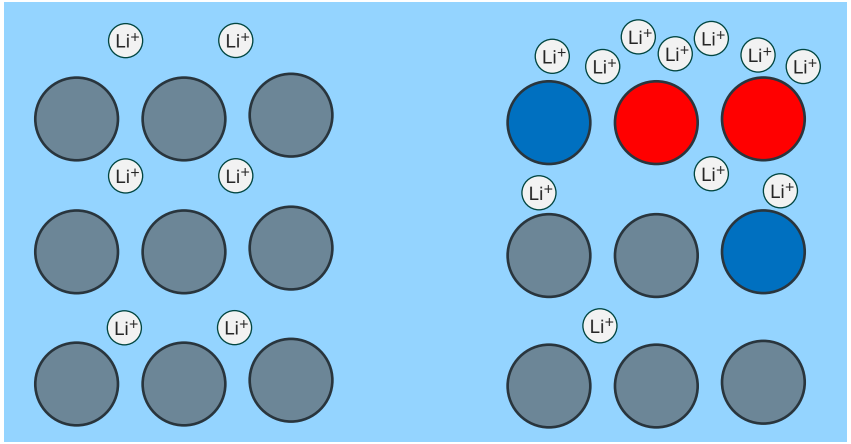
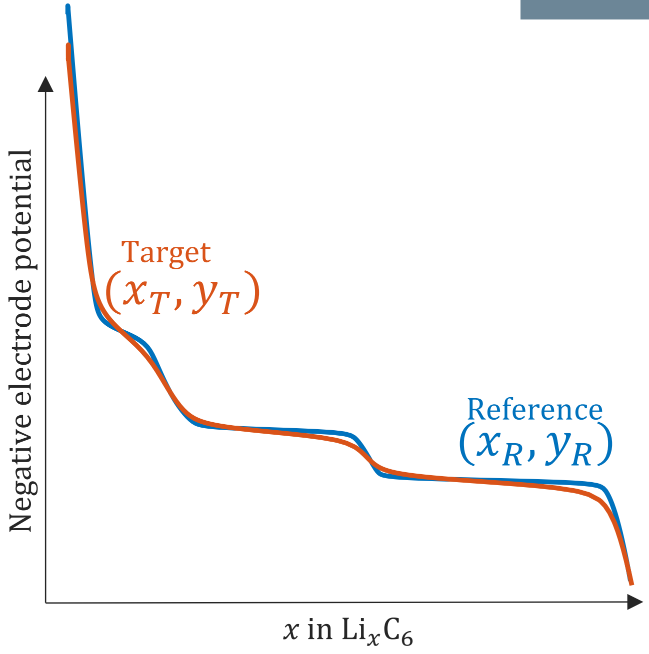
Estimation at higher currents

At higher currents:

- Transport limitations lead to uneven charge of electrode particles
- This leads to a smoothing of the electrode voltage, since it is an average over all particles

Reference (slow) Target (faster)





Estimation at higher currents

KDF: $f_{\mu\sigma}$, parameters μ, σ: logit-normal

Solve for each x_T, y_T

$$\min_{\sigma_t} \sum_{k=1}^{N} \lambda_k^{|t-k|} \left(\mathbb{E}_{f_{x_T(k)\sigma_t}}[y_R] - y_T(k)\right)^2,$$

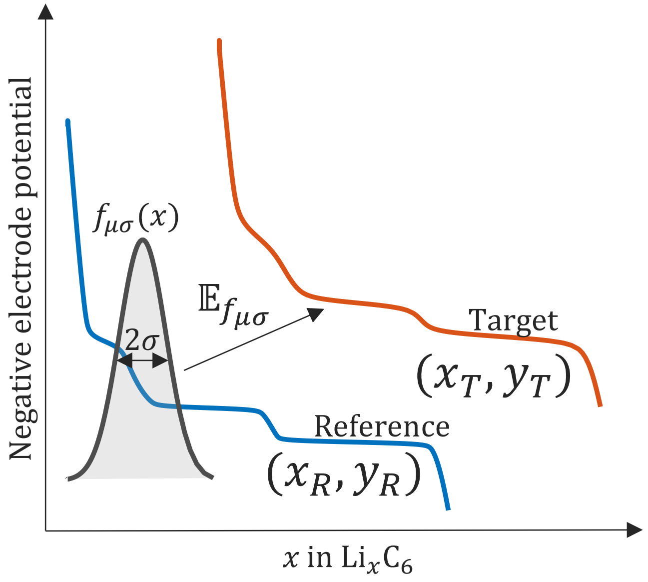
$$\text{s.t } \sigma_t > 0,$$

$$\mathbb{E}_{f_{\mu\sigma}}[y_R] = \int f_{\mu\sigma}(x_R)\, y_R(x_R) dx_R$$

Forgetting factor $\lambda_k \in (0,1)$

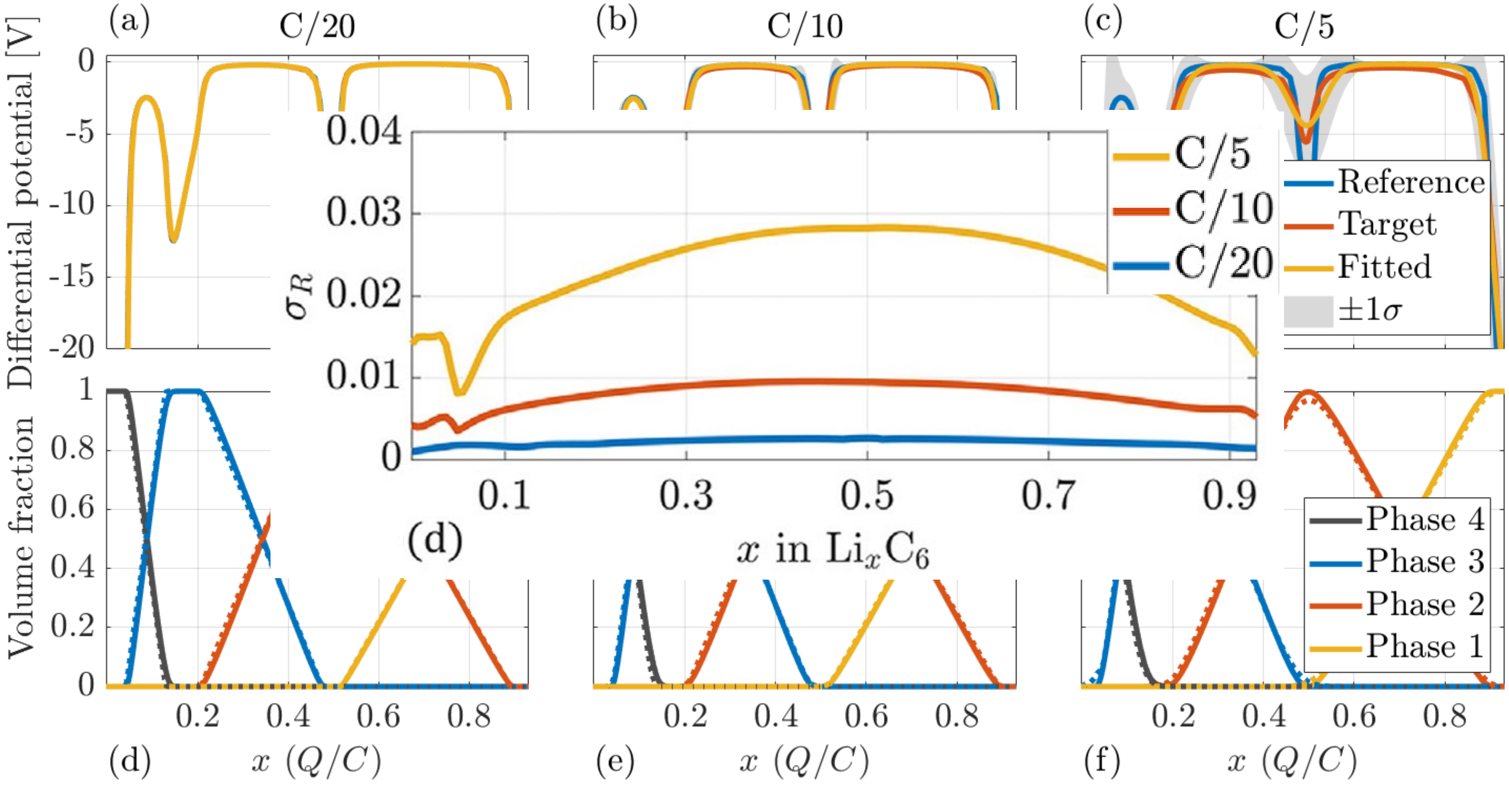
Given σ, estimated weight fraction is

$$\hat{p}_T^i = \mathbb{E}_{f_{\mu\sigma}}\left[\hat{p}_R^i\right], \qquad i = 1, \ldots, 4$$

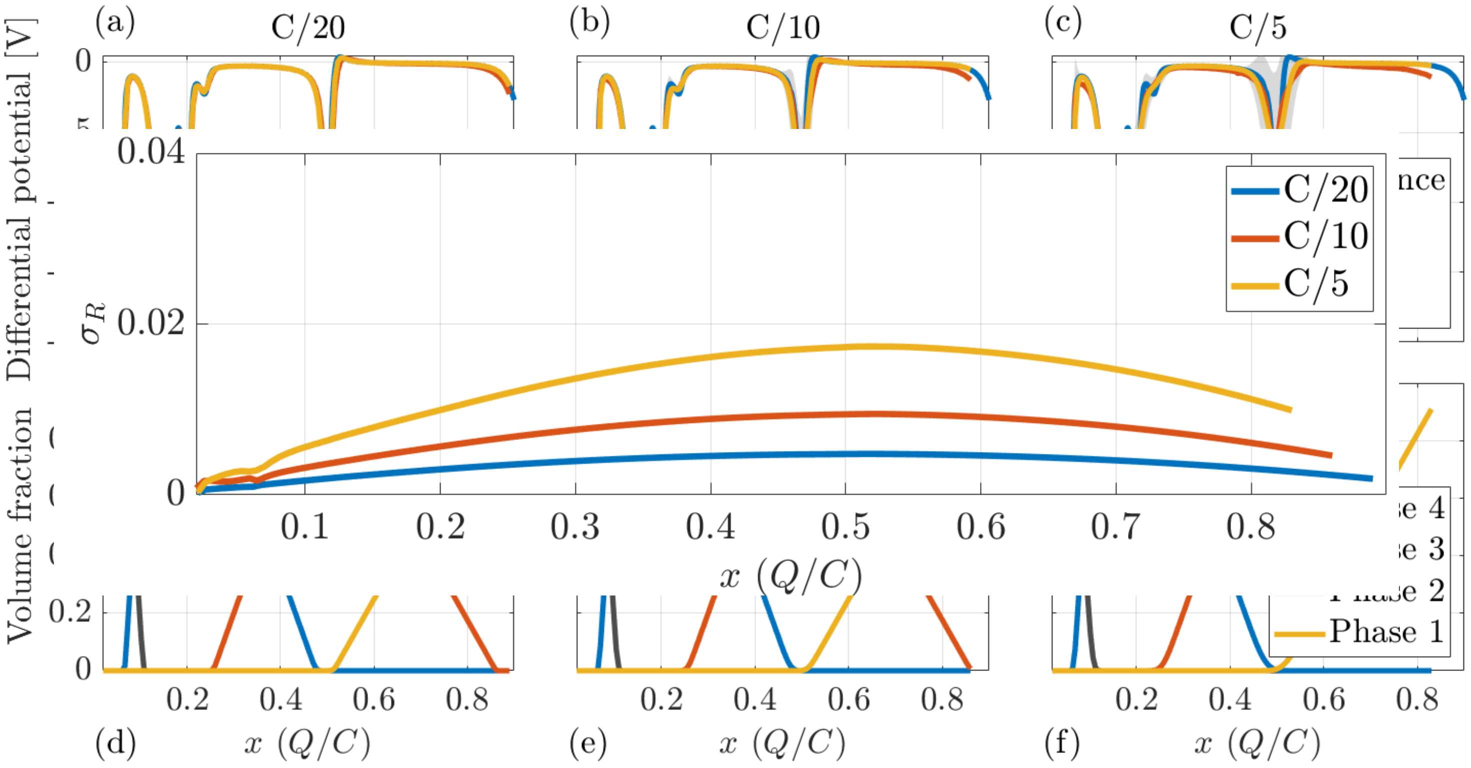


The value of σ does not only depend on current: material properties affect it → can possibly diagnose manufacturing process

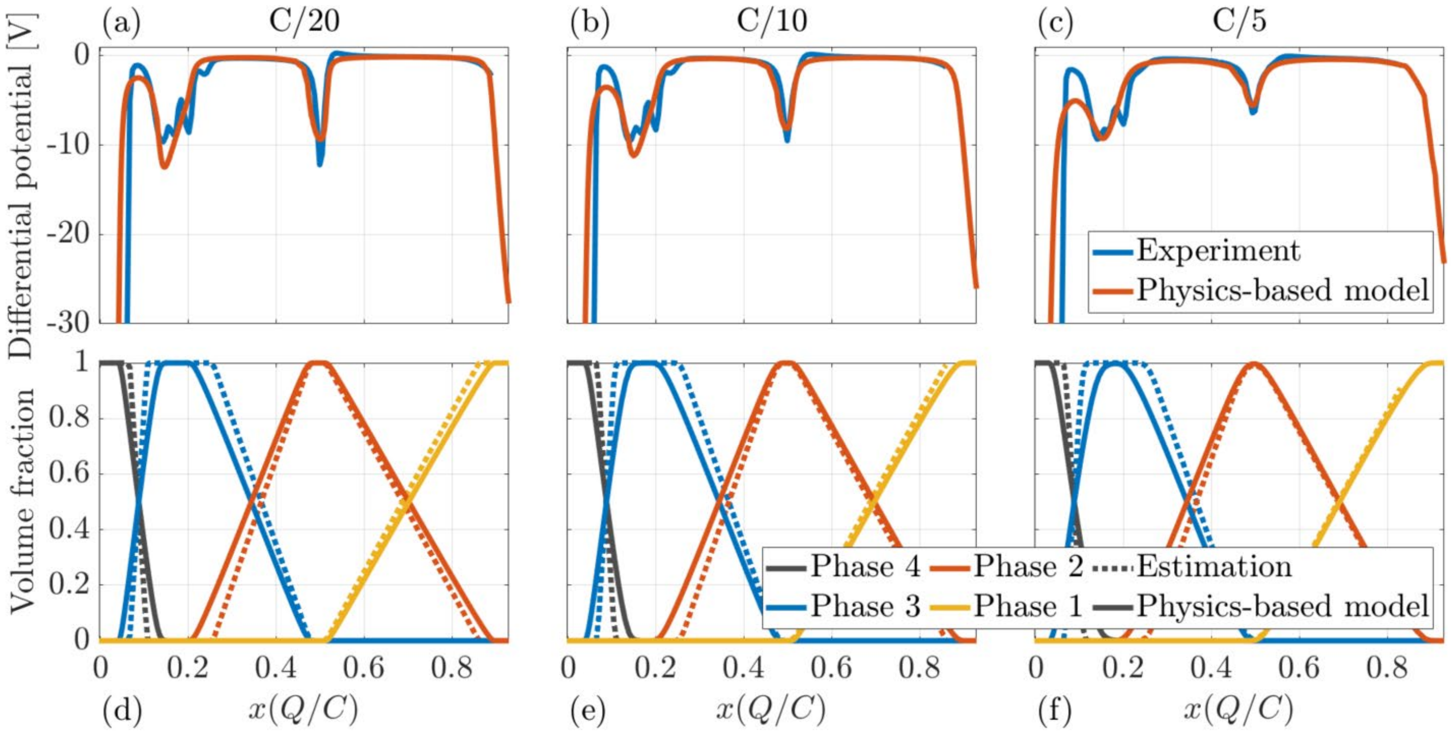
Simulation results [ref I=C/40]



Experimental results [ref l=C/40]



Results comparison



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Discussion & conclusions

- Works well for half cells:
  - ICA knee-points align with phase peaks at low current
  - Kernel smoothing extends phase estimation to higher currents
- Good SOC estimation is **critical**
  - X-axis must align between target & reference
- Could be used as diagnosis tool in battery manufacturing
  - Fitted σ depends on material properties
- Before adaption to full cells: PE must be accounted for
  → Higher uncertainty
- Adapt method to varying current scenarios, validate for higher currents
  - Would be needed for feedback

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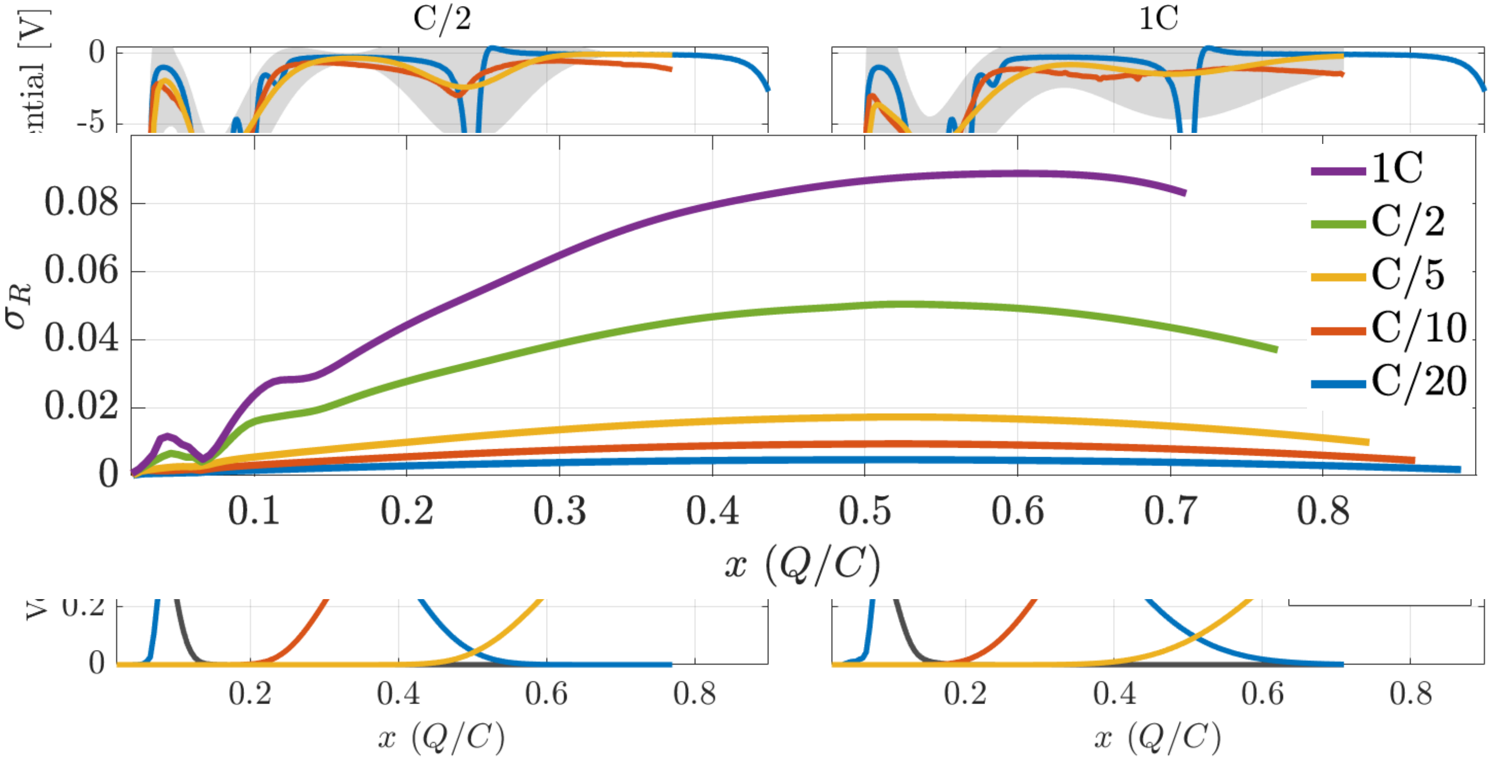
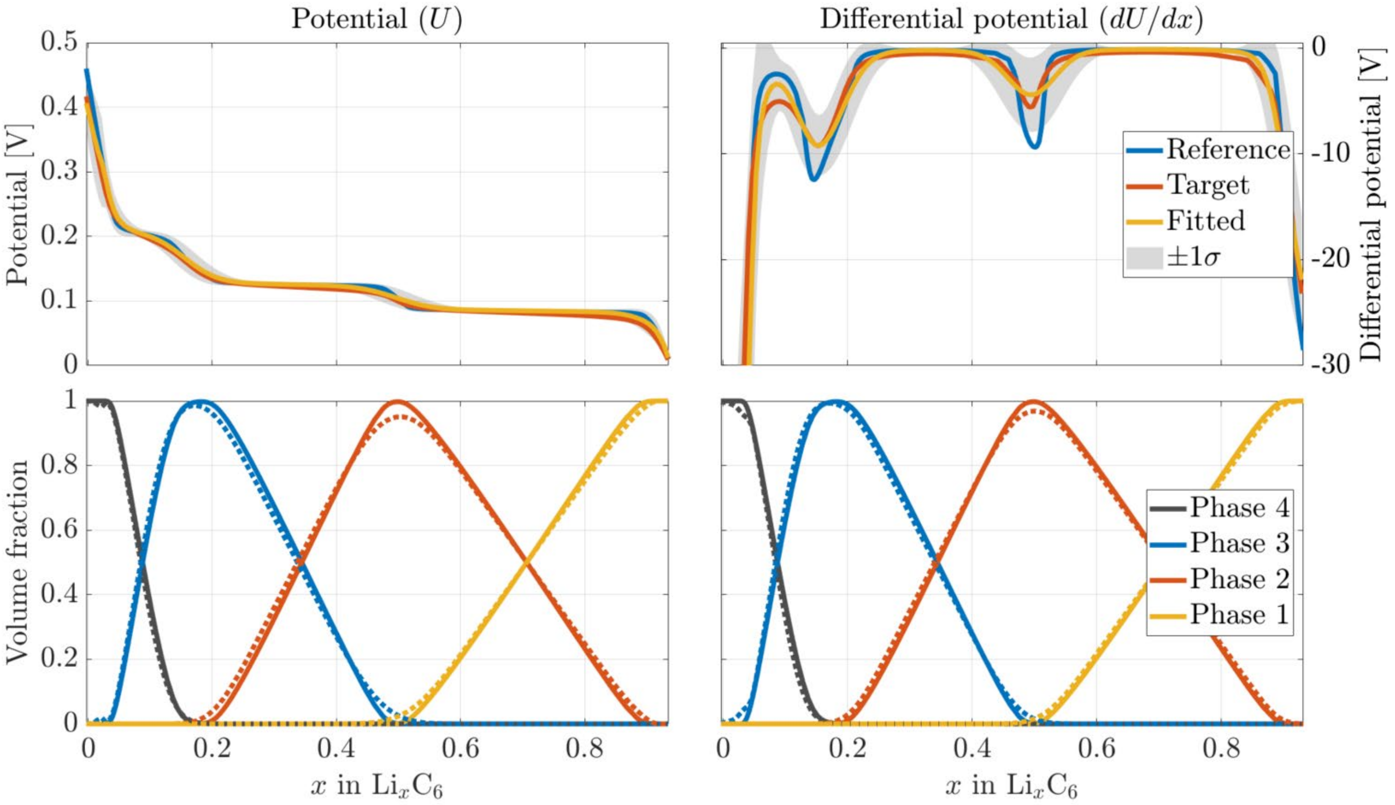
Experimental results [ref I=C/40]



Simulation results [I=C/5, ref I=C/40]



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Fitting results

	Mean σ		Normalized MAE y		MAE p
I [Target]	Simulation	Experiment	Simulation	Experiment	Simulation
C/20	0.00214	0.00350	0.0313	0.0469	0.0192
C/10	0.00790	0.00694	0.0775	0.0604	0.0234
C/5	0.0229	0.0127	0.140	0.0616	0.0337
C/2	-	0.0369	-	0.139	-
1C	-	0.0633	-	0.158	-

Phase reference MAE (sim): 0.0192