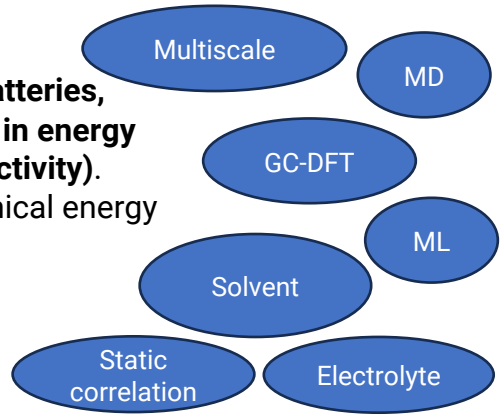
Christopher Ehlert | 14.12.2023

# Challenges in Modelling of Electrochemical Energy Materials

## Introduction [1]

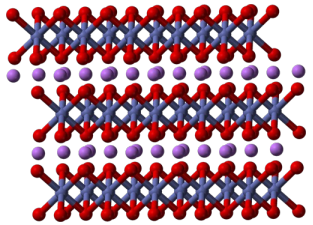



**Materials involved in energy storage systems (batteries, supercapacitors)** and **electrochemical interfaces in energy conversions ($CO_2$RR, ORR, Fuel Cells → local reactivity)**.
Goal: understand, control and design electrochemical energy materials at atomistic precision.
What makes this field challenging?:

- Solid-Liquid interfaces.
- Interfaces are charged and "electrified".
- Electrons and Ions.

## Modelling and characterisation of transition metal oxide electrodes [1]

Lithium transition metal oxides (TMOs) have the general formula $Li_xM_yO_z$
NMCs $LiNi_xMn_yCo_zO_2$ discovered in 2001, now important cathode material in electric vehicles. NMC532 → $LiNi_{0.5}Mn_{0.3}Co_{0.2}O_2$
3 general forms: layered, spinel, disordered rocksalt (DRX)



**We need:**

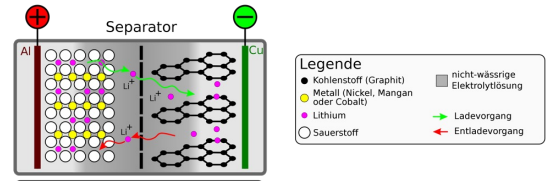
- high gravimetric and/or volumetric energy density
- high rate capability
- stability for safety and long-term cyclability
- low cost of production

**Challenge**:

- Layered NMCs have high energy density, good stability but utilise Co. Remove Co → degrade stability.
- **Understanding the degradation mechanisms (at different Li loadings).**
- **Understanding changes in oxidation states during cycling.**

**First principles** studies to understand impact on mechanical properties (elastic modulus, hardness) **[2,3]**
Li is extracted from NMC materials: Jahn–Teller distortion, depletion of electrostatic interactions of Li–O, and weak ionic: TM–O bonding contribute to the change in mechanical properties.

Oxidation states are accessible through X-ray absorption → Interplay with theory.
**Future:** intersecting modelling, characterization, and ML

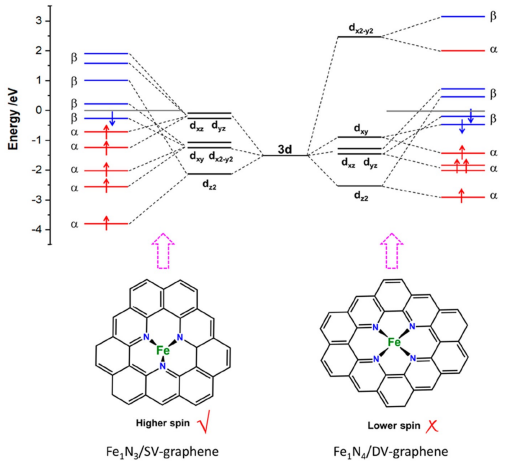



## Theoretical understanding of single-atom electrocatalysis [1]

*"Single atom catalysis describes a process in which a single atom on a catalyst surface drives a catalytic reaction. The catalyst with a single atom on its surface is called a single atom catalyst (SAC)."*
**Example**: $Pt_1/FeO_x$ much better in CO oxidation reaction that a Pt nanoparticle.
Important for: HER, OER, ORR, $CO_2$RR.
SAEC: Single atom electrocatalysts involve an electrode+applied potential.

**Challenge:** active sites of SAECs are often composed of transition metal atoms with strong static correlation → DFT may break, but CASSCF & MRCI are too expensive.
Computational Hydrogen Electrode model (CHE): simplifies the influence of an electrode potential. An explicit electrode potential:

- can change underlying mechanisms dramatically.
- can induce a dynamic transformation of active site structure.
- can induce degradation of SAECs.






**We need:**

- Generate (electrode) potential-dependent reaction networks.
- Quantum embedding might be useful [1].
- ML for "new" descriptors.

**[1]** *J. Phys. Energy*, **2023**, *5*, 041501. **[2]** *Phys. Chem. Chem. Phys.* **2017**, *19*, 1762. **[3]** *J. Electrochem. Soc.* **2017**, *164*, A3333.