# Electro-chemo-thermo-mechanical Coupled Model for Lithium-ion Batteries

**Yitao Qiu[1], Xiaoxuan Zhang[1,2], Sergei Chumakov[3], Markus Klinsmann[4], Daniel Mayer[3], Xiaobai Li[3], Sun Ung Kim[5], Jake Christensen[3] and Christian Linder[1]**

[1] Stanford University, Department of Civil & Environmental Engg. [2] University of Michigan, Department of Mechanical Engg.
[3] Robert Bosch LLC, Research & Technology Center [4] Robert Bosch GmbH, Corporate Research [5] Washington State University, School of Engg. & Computer Science

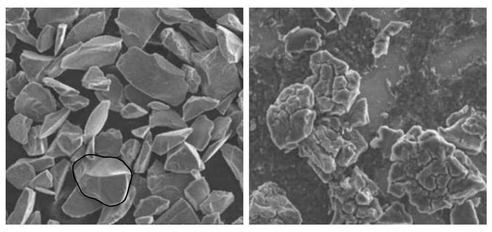
## Introduction

Lithium-ion batteries are important energy storage devices with a wide range of applications. Developing advanced multi-physics models to describe the complex physical processes happening in batteries is crucial for understanding different aging mechanisms, improving cell design, and better controlling cells in a battery management system. In addition, for battery modules or packs with larger size, the inhomogeneity of the electric and the thermal fields exhibited by the spatial scale greatly affects the performance of the battery.

portable electronics

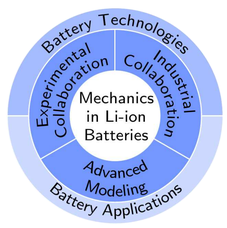
electric vehicle


stationary energy storage



microscale: Si nanoparticles before/after charging [1]


macroscale: an aged 20 Ah LiNiMnCo cell [2]




overview of LIBs research

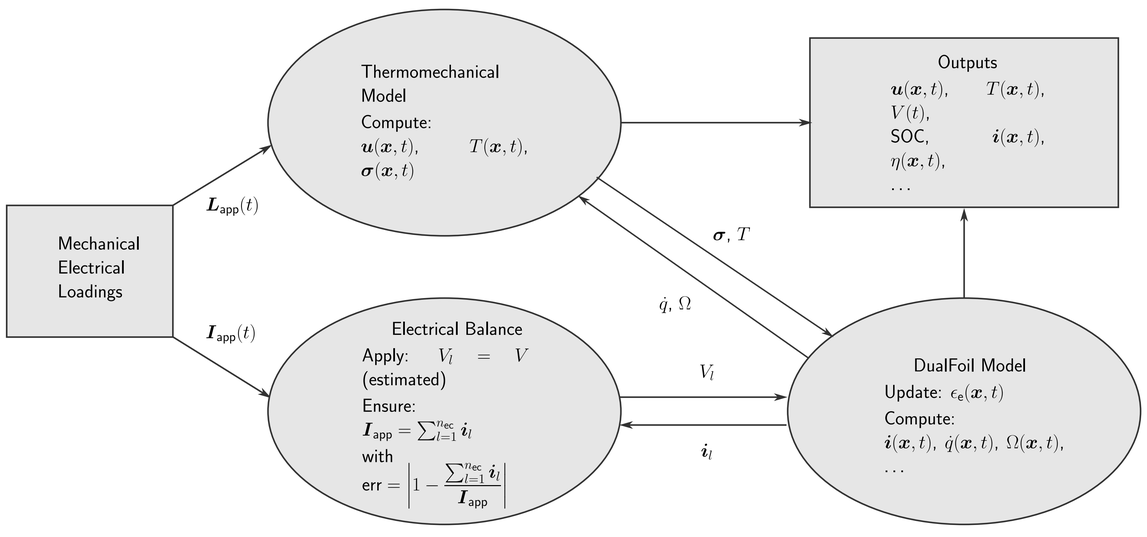
In this work, we developed an electro-chemo-thermo-mechanical coupled model to study how mechanical deformation affects the electrochemical performance of lithium-ion batteries. In this model, an electro-chemical (so-called DualFoil) model is coupled with a thermo-mechanical model. Mechanical deformation impacts cell electro-chemical properties via the porosity change in the electrodes and the separator due to interrelation induced volume change of the active materials and externally applied mechanical loading. The model can be used to efficiently study 3D cells with large geometry and resolve the spatial variation of the fields of interest. Furthermore, the correlation between mechanical deformation and Lithium plating can be explained using this model.

## Theory

| Equation description | Equation |
|---|---|
| Electrolyte material balance | $\frac{\partial(\epsilon_e c_e)}{\partial t} = \nabla_x \cdot \left( D_{e,\text{eff}} \nabla_x c_e + \frac{\boldsymbol{i}_e (1 - t_+)}{F} \right)$ |
| Electrolyte-phase Ohm's law | $\boldsymbol{i}_e = -\kappa_{e,\text{eff}} \nabla_x \phi_e + 2\frac{\kappa_{e,\text{eff}} R\theta}{F}(1 - t_+)\left(1 + \frac{\partial \ln f_\pm}{\partial \ln c_e}\right) \nabla_x \ln c_e$ |
| Intercalate material balance | $\frac{d}{dt}\bar{c} + 3\frac{j_n}{R_p} = 0$ |
| Volume-averaged flux relation | $\frac{d}{dt}\bar{q} + 30\frac{D_s}{R_p^2}\bar{q} + \frac{45}{2}\frac{j_n}{R_p^2} = 0$ |
| Intercalate boundary conditions | $35\frac{D_s}{R_p}(c_s - \bar{c}) - 8 D_s \bar{q} = -j_n$ |
| Solid-phase Ohm's law | $\boldsymbol{i}_s = -\kappa_{s,\text{eff}} \nabla_x \phi_s$ |
| Butler-Volmer insertion kinetics | $j_n = k_0 c_s^{\alpha_c} c_e^{\alpha_a} \left(c_s^{\max} - c_s\right)^{\alpha_a} \left[\exp\left(\frac{\alpha_a F}{R\theta}\eta_s\right) - \exp\left(-\frac{\alpha_c F}{R\theta}\eta_s\right)\right]$ with $\eta_s = \phi_s - \phi_e - U_s$ |
| Charge conservation | $\nabla_x \cdot (\boldsymbol{i}_s + \boldsymbol{i}_e) = 0$ with $\nabla_x \cdot \boldsymbol{i}_e = -aF j_n$ |
| Balance of linear momentum | $\text{Div}[\boldsymbol{P}] = \boldsymbol{0}$ with $\boldsymbol{F} = \boldsymbol{F}_{\text{elastic}} \boldsymbol{F}_{\text{swelling}} \boldsymbol{F}_\theta$ |
| Energy balance equation | $\rho C_p \frac{\partial \theta}{\partial t} + \nabla \cdot (-\kappa_\theta \nabla \theta) = \dot{q}$ |

Table: Summary of equations used to describe the involved physics in the model [4, 5].

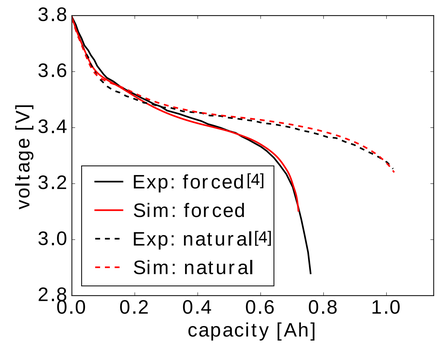
## Simulation Procedure

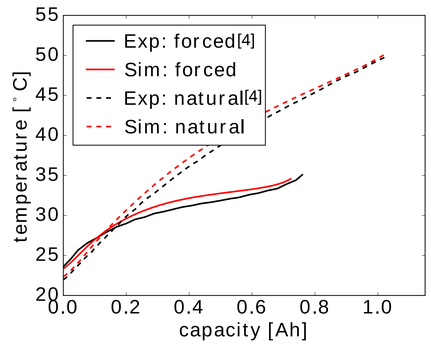


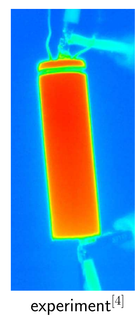

- Thermomechanical Model: A coupled model that solves for the displacement and temperature fields at the large deformation setting.
- DualFoil Model: An electrochemical model that describes the charge-transfer kinetics, mass transport, and the electric potential variations inside a battery.
- Electrical Balance: An user-defined API to apply the electrical loading.
- Li Plating: Its occurrence is related to the anode surface potential, $\eta_a = \phi_s - \phi_e - U_{\text{Li}}$.

## Numerical Examples

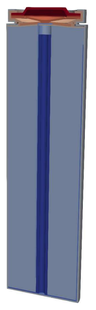
- Discharge simulation of a commercial 18650 cell



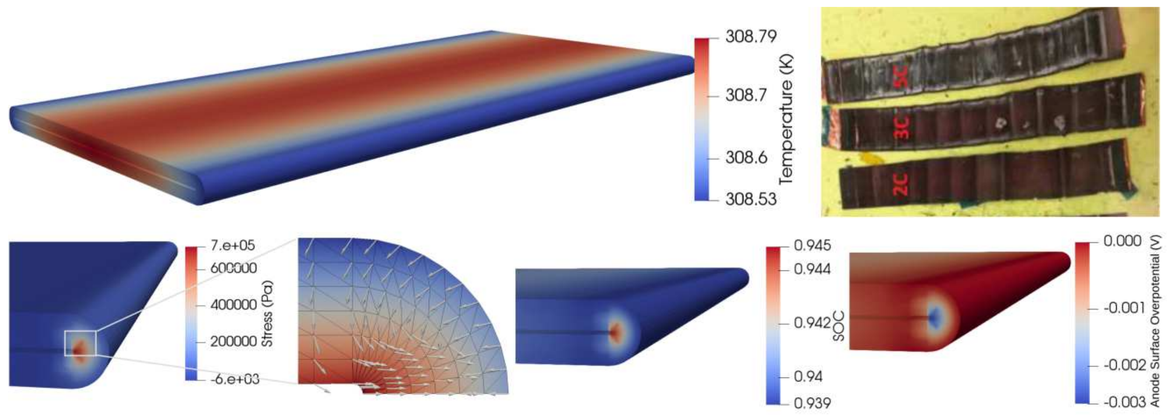



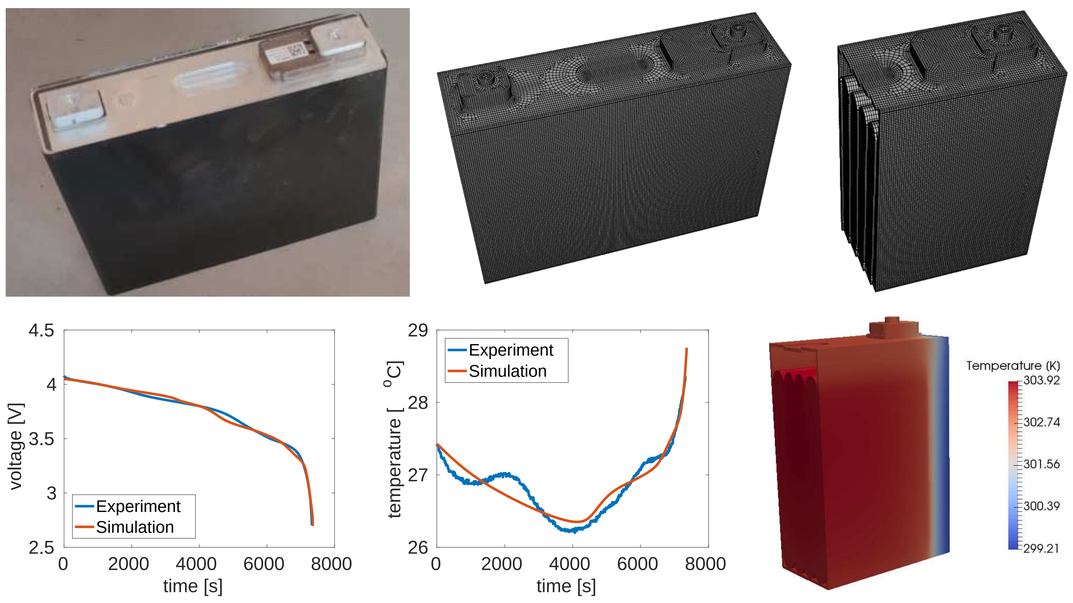
experiment[4]



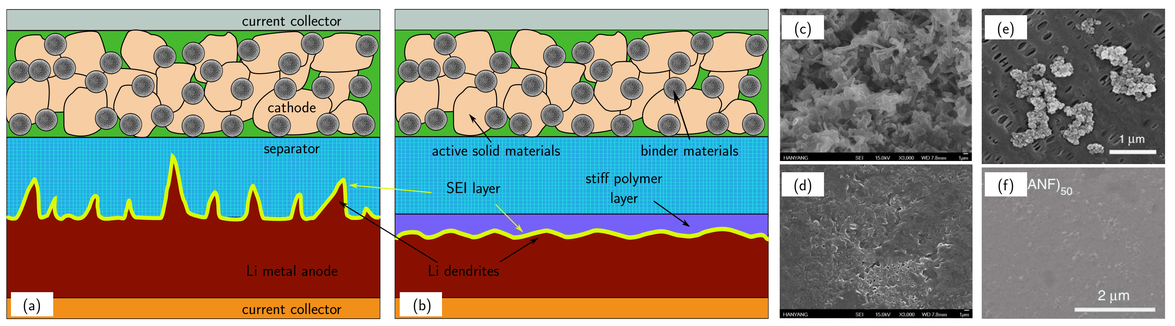
- Charge/discharge simulation of a prismatic cell




- Discharge simulation of a high capacity electric vehicle cell




## Future Research




- Li metal is an ideal anode material because of its high theoretical specific capacity, low density, and the lowest negative electrochemical potential.
- Dendritic Li growth poses a major safety challenge to Li metal based cells.
- Mechanical constraints have been demonstrated both theoretically and experimentally to be able to suppress Li dendrite formations, as shown in Figure (c)-(f) [6-8].
- Goal: Develop novel multiphysics computational tools to understand mechanical impact on Li dendrite formation mechanism.

## Funding Supports

This work was sponsored in part by Robert Bosch LLC through Bosch Energy Research Network (BERN) Grant No. 01.01.MS.17.

## References

[1] K. Rhodes et al. (2010). *J. Electrochem. Soc.* 157:A1354-A1360.
[2] C. Birkenmaier et al. (2015). *J. Electrochem. Soc.* 162(14):A2646-A2650.
[3] X. Zhang, A. Krischok & C. Linder (2016). *Comput. Methods Appl. Mech. Engrg.* 312:51-77.
[4] J. Christensen, D. Cook & P. Albertus (2013). *J. Electrochem. Soc.* 160(11):A2258-A2267.
[5] X. Zhang et al. (2019). *J. Power Sources* 412:552-558.
[6] C. Monroe & J. Newman (2003). *J. Electrochem. Soc.* 150(10):A1377-A1384.
[7] X. Zhang et al. (2020). *J. Electrochem. Soc.* 167(16):160542.
[8] X. Zhang et al. (2021). *J. Electrochem. Soc.* 168(2):020533.
[9] Y. Qiu et al. (2022). *J. Power Sources* 541:231632.

Stanford | ENGINEERING
Civil & Environmental Engineering

Stanford University

Stanford | DOERR
School of Sustainability