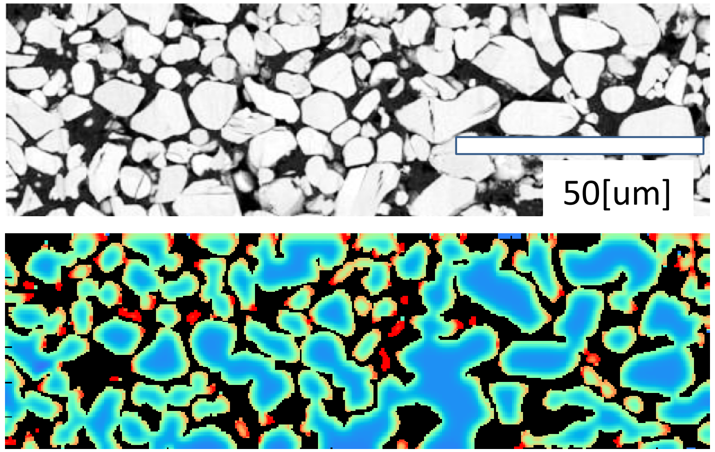
ver. 2020-8



KOBE STEEL GROUP

Battery Simulation Using COMSOL Multiphysics

Kobelco Research Institute Inc.

From analysis, testing, and measurement to manufacturing and related testing, Kobelco Research supports customer businesses through a variety of approaches.

Company profile

Kobelco Research Institute Inc.

Affiliated by KOBE STEEL, LTD

Business: Analysis, testing, prototyping, supporting R&D

Employees: 1366 (including 58 Ph.Ds)

Headquarters: Kobe, Japan

Analysis

Support for solving R&D and manufacturing problems in all types of industries

The analytical and technical strength of Kobelco Research provides support for research, development, and manufacturing problems in all types of industries addressing materials, organic, environmental, and physical analysis.

Prototype / Experiments

Support for new product and new process R&D, including ferrous and non-ferrous materials

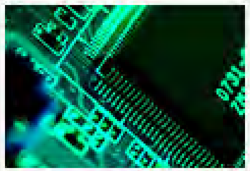
Services performed include clean room, soil, and other environmental testing, materials-related stress measurement, and non-destructive testing (detection and evaluation of surface and internal defects in materials and structures, integrity and quality evaluation, and detection and evaluation of materials damage and degradation).

Testing

Support for R&D work as industry structures change

As industry structures change, demand for new corrosion-resistant materials and surface treatment technologies is increasing. When new technology development in these areas is needed, the research and development work itself can be performed on a contract basis.

Manufacturing

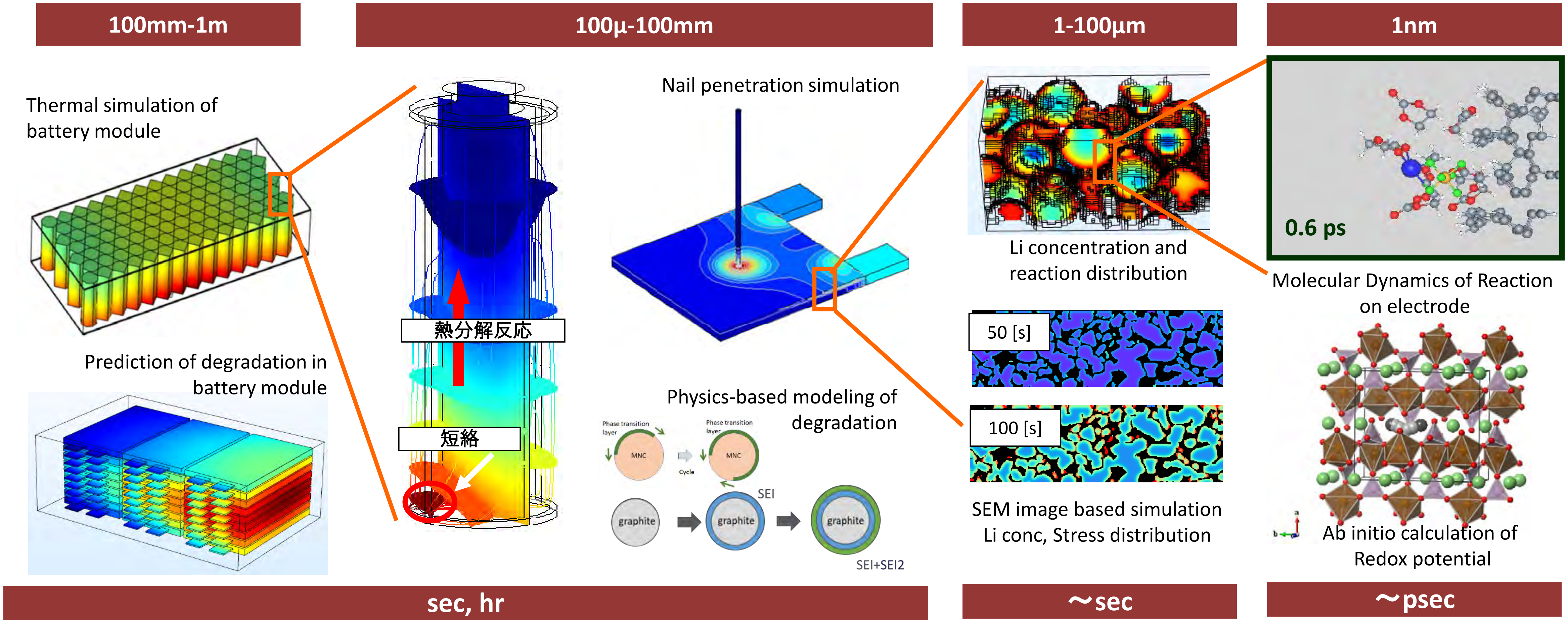


Manufacturing semiconductor-related materials , testing systems , medical materials and PV-related parts

We manufacture and market

- Sputtering target materials
- Semiconductor testing systems
- Co-Cr tubes for stent
- Metallic products coated with carbon-based thin film
- Screen for PV cell

Simulation Technologies for Li-ion battery



Safety evaluation

Operation condition

Degradation prediction

Optimization
Inverse problem

Materials Informatics

Machine Learning, Data analysis

Topics

- Equivalent Circuit Model for Battery Management System
- Physico-chemical Simulation using FIB-SEM image
- Battery Safety Simulations, Nail penetration test, Burning test
- Battery Degradation Simulation
- Machine Learning, Deep Learning

Topics

- ☐ Equivalent Circuit Model for Battery Management System
- ☐ Physico-chemical Simulation using FIB-SEM image
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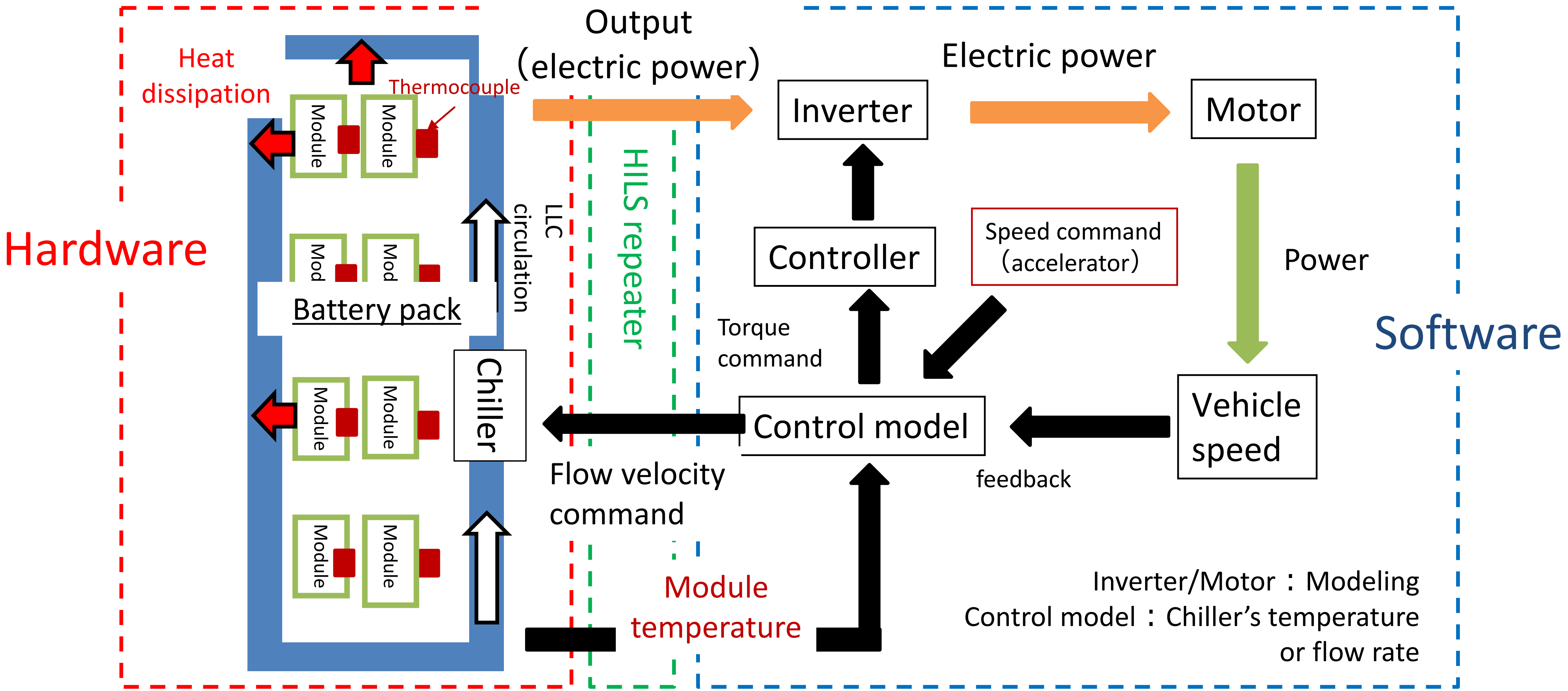
- ☐ Machine Learning, Deep Learning

Battery charge/discharge test in EV virtual driving environment

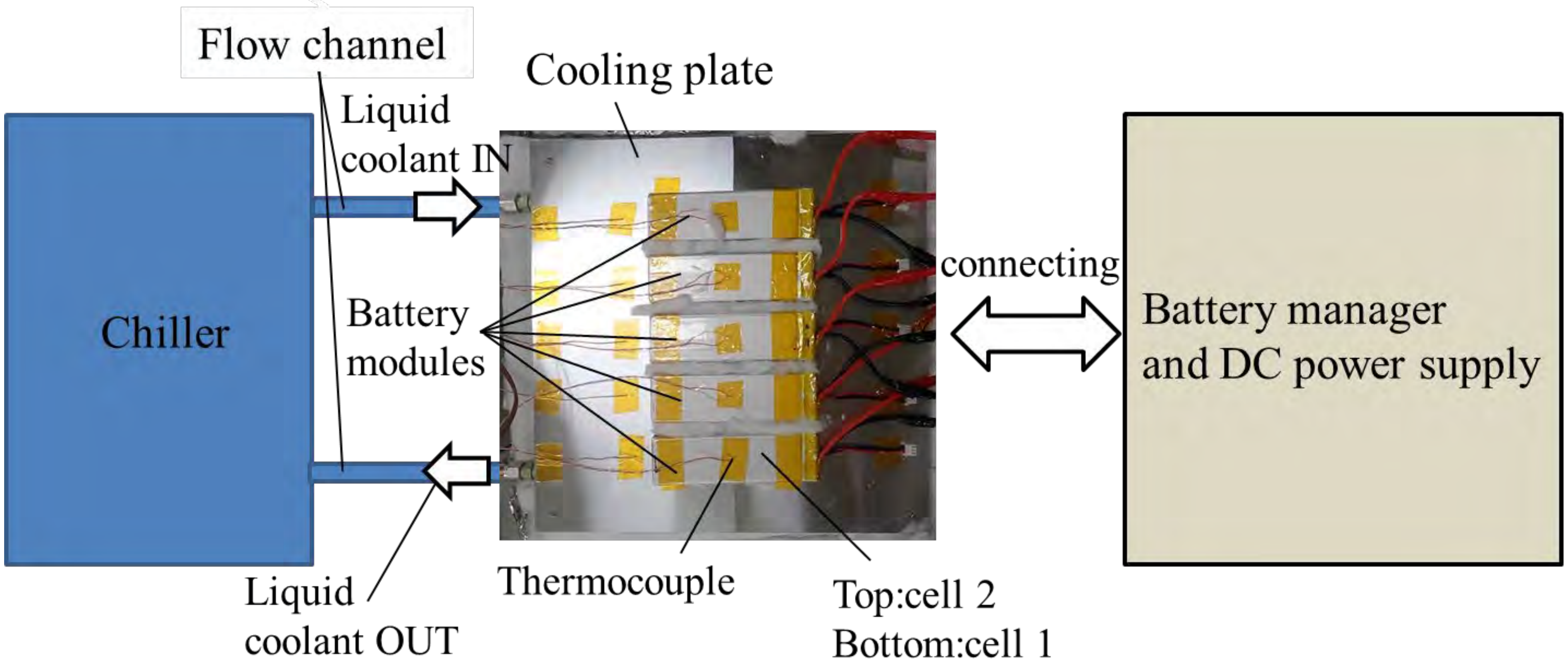


✓Reproduce the load on the battery under the assumed driving conditions.
✓The control algorithm of the battery cooling device can be examined.

KOBELCO RESEARCH INSTITUTE, INC.

Equivalent Circuit Model for Battery Management System

✓Construction of an equivalent circuit model for battery management systems is shown.
✓Test bench assuming an actual cooling system is constructed.



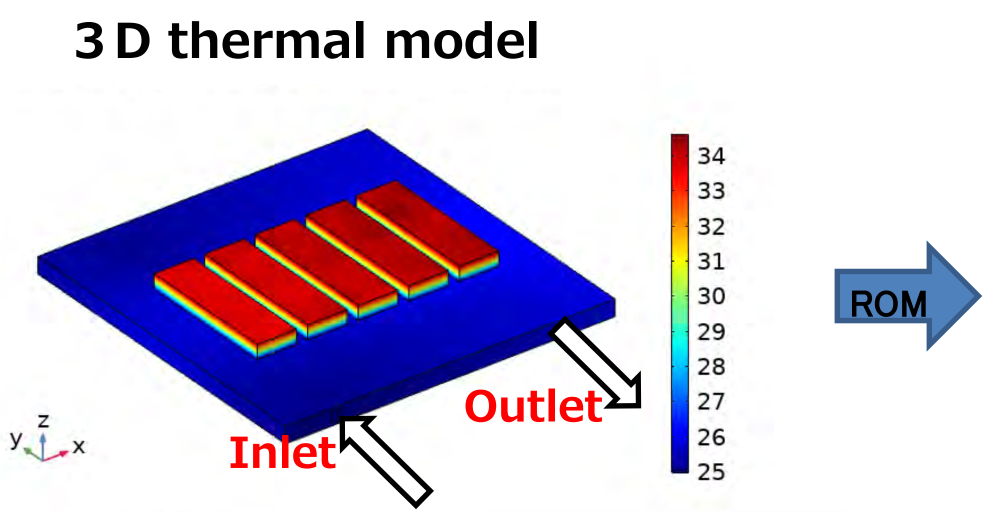
<u>Test conditions</u>

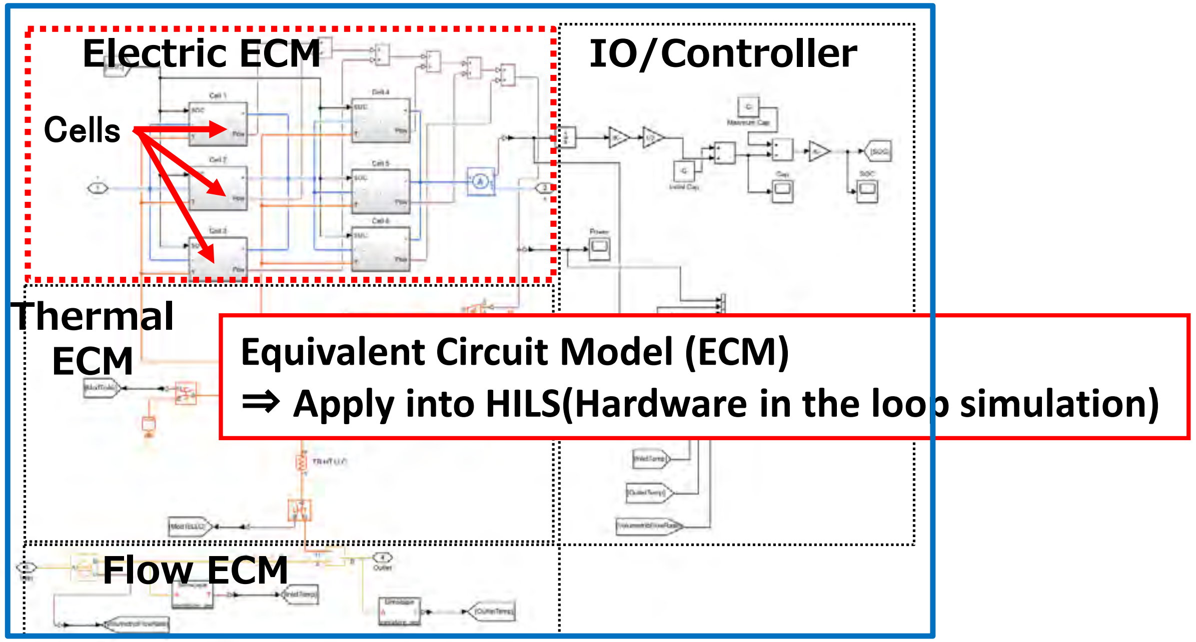
・2 current rate CC charge / discharge × 3 cycle without the flow
・2 current rate CC charge / discharge × 3 cycle with the flow 1.0 l/min

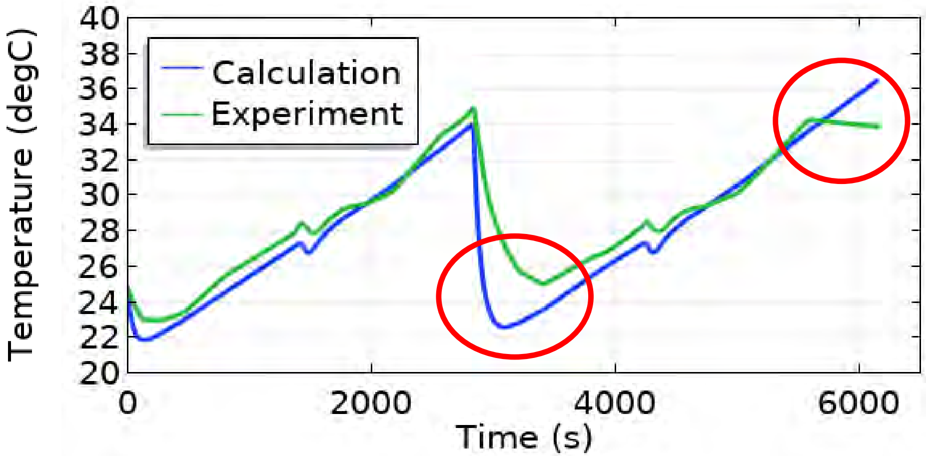
Equivalent Circuit Model for Battery Management System

Real-time simulation with equivalent circuit models

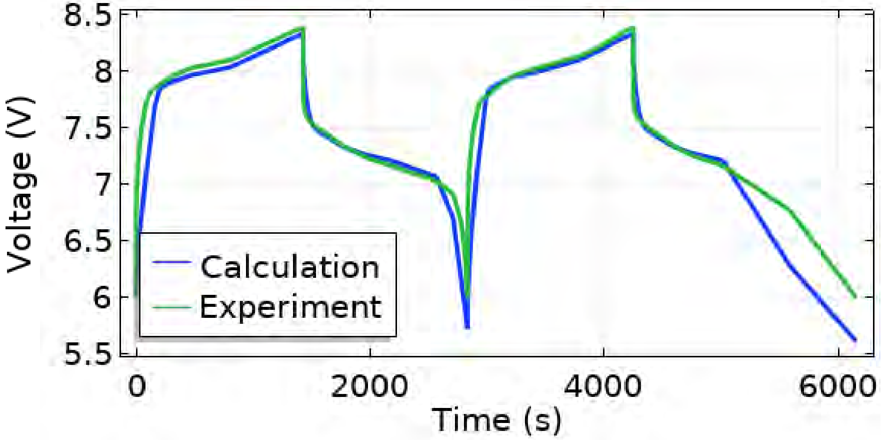
T. Yamanaka *et al*., Batteries, **2020**, *6*(3), 44.







Comparison for time histories of temperature



Comparison for time histories of voltage

Topics

- ☐ Equivalent Circuit Model for Battery Management System
- ☐ Physico-chemical Simulation using FIB-SEM image
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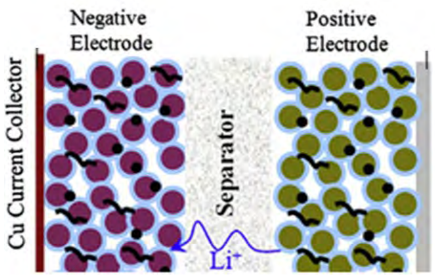
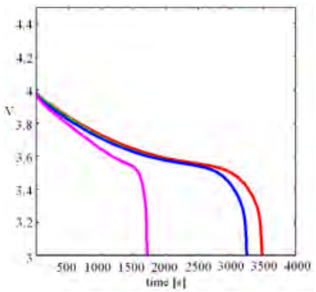
- ☐ Battery Degradation Simulation
- ☐ Machine Learning, Deep Learning

Physico-chemical Simulation using FIB-SEM image

Conventional: 1-D Newman model





S.J. Harris et al. / Chemical Physics Letters 485 (2010) 265.

Physico-chemical model

Electrode potential:
Poisson Equation

$$\mathbf{i}_s = -\sigma_s \nabla \phi_s$$

Electrolyte potential :
Nernst-Plank Equation

$$\nabla \cdot \left(-\sigma_l \nabla \phi_l + \frac{2\sigma_l RT}{F}\left(1 + \frac{\partial \ln f}{\partial \ln c_l}\right)(1 - t_+)\nabla \ln c_l \right) = i_{tot}$$

Li conc. in AP

$$\frac{\partial c_s}{\partial t} = \nabla \cdot (-D_s \nabla c_s)$$

Electrochemical reaction:
Butler-Volmer Equation

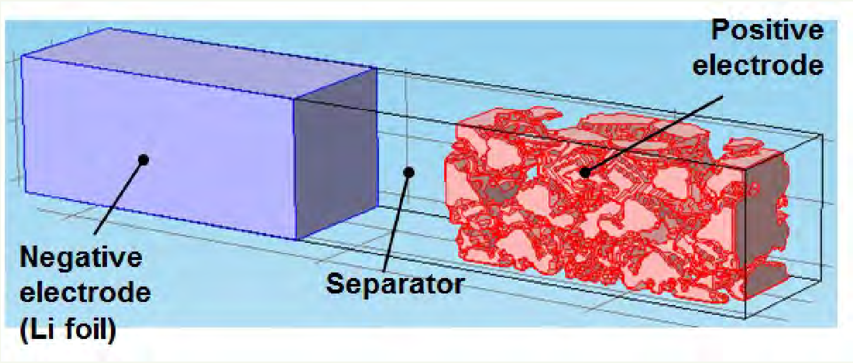
$$i_{loc} = i_0\left(\exp\left(\frac{\alpha_a F \eta}{RT}\right) - \exp\left(\frac{-\alpha_c F \eta}{RT}\right)\right)$$

Quasi-3D modeling using FIB-SEM image

(～100[um])

Multiphysics Simulation based on 3D microstructure

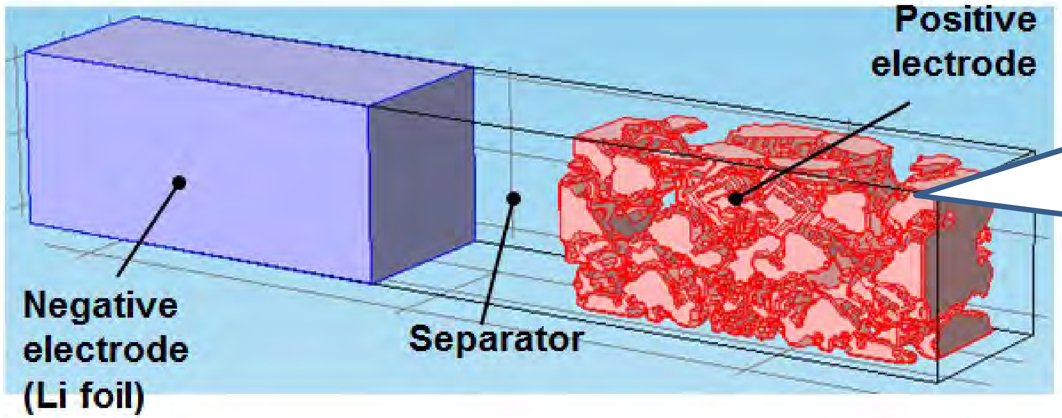
(～50[um])

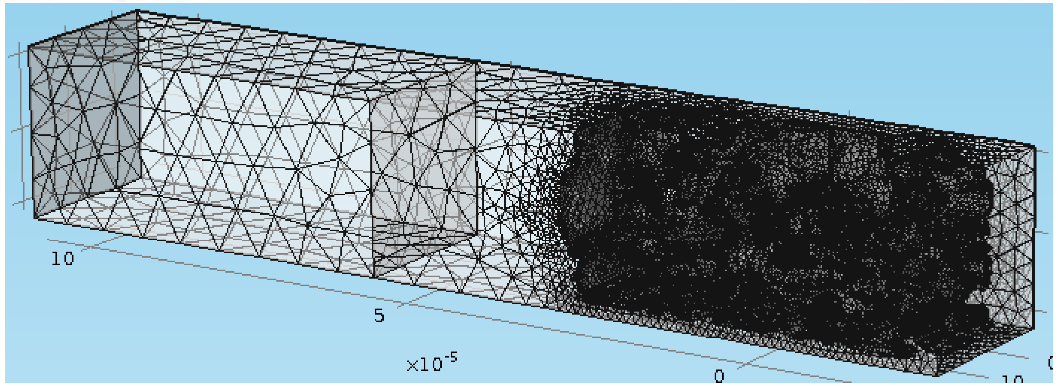


Multiphysics Simulation based on 3D microstructure

Charge/discharge FEM simulation based on 3D microstructure

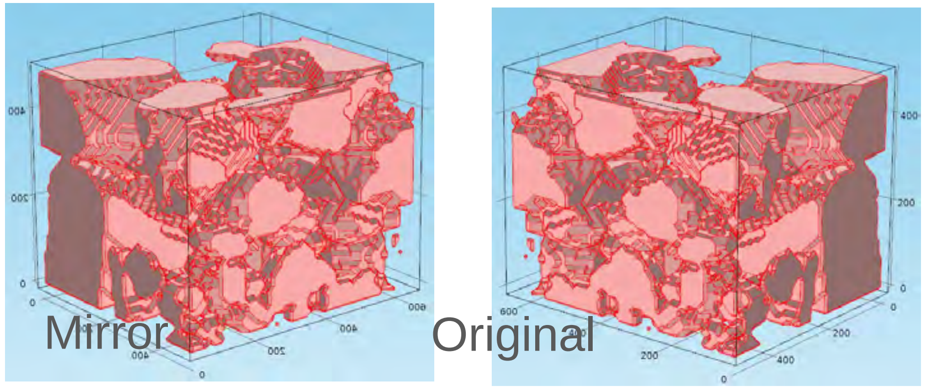
Model geometry





Tetra mesh 1million elements

Positive electrode structure



Thickness, positive electrode: 50 [um]

Thickness, negative electrode (Li foil)：50 [um]

Thickness, separator：25 [um]

OCP, positive：Measurement (NMC1/3)

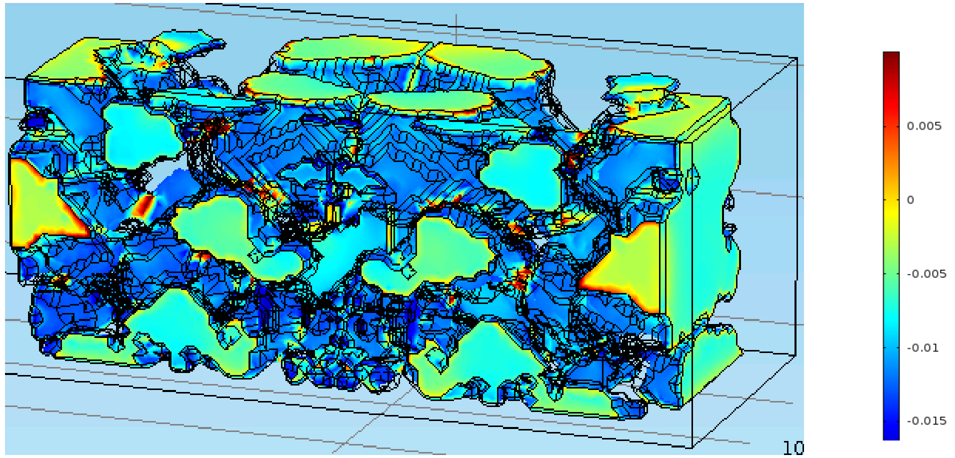
Reaction rate coefficient：1.0e-11 (assumed)

Li diffusion coefficient：1.0e-14（assumed）

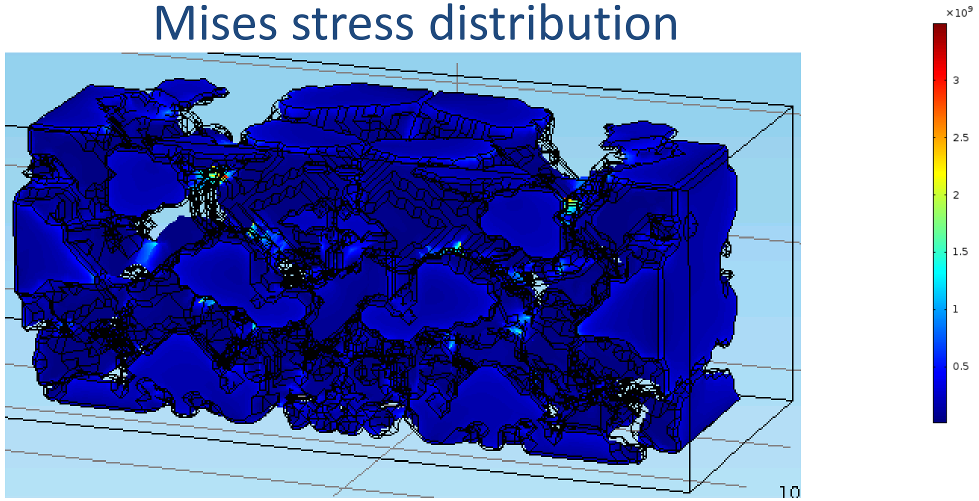
Multiphysics Simulation based on 3D microstructure

Charge/discharge FEM simulation based on 3D microstructure

Strain distribution during discharge



Mises stress distribution



Stress balance

$$\sigma = C(\varepsilon - \varepsilon_L)$$

Eigen strain

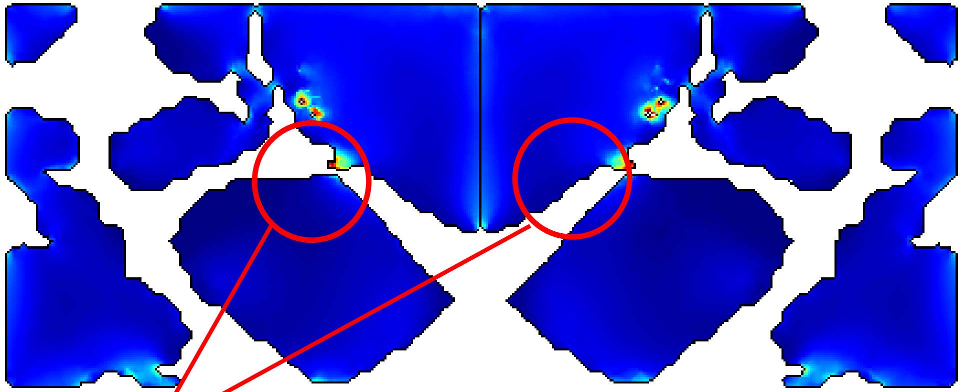
$$\varepsilon_L = \frac{\varepsilon_{\max}}{c_{\max}} c$$

Young's modulus: 70 [GPa] *1

Poisson ratio: 0.3 (assumed)

Volume change coefficient: 0.03 *2

Slice image of positive electrode

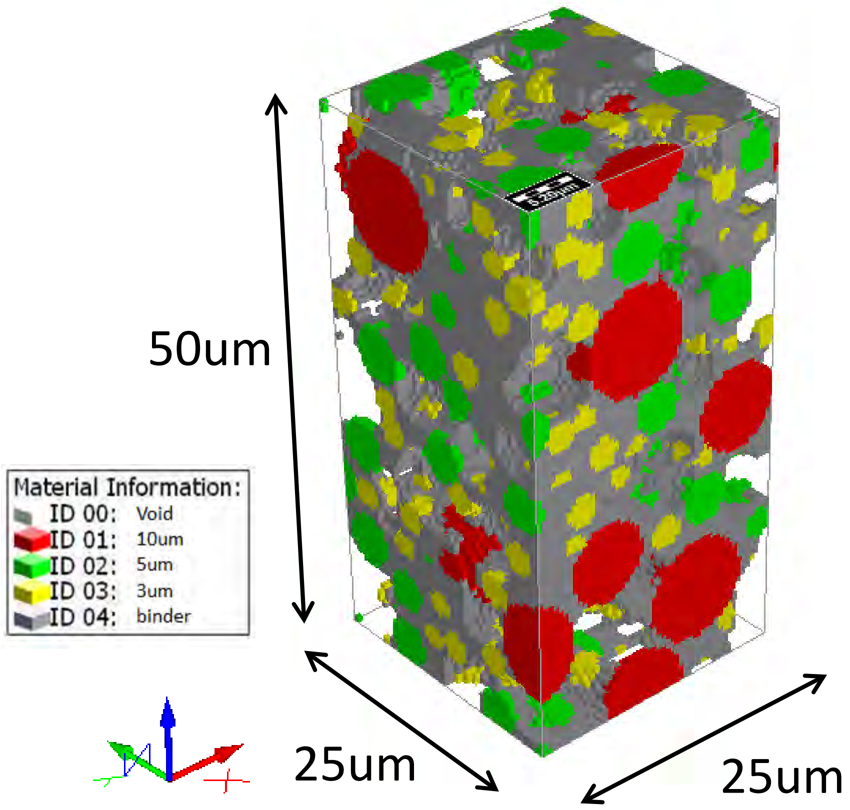


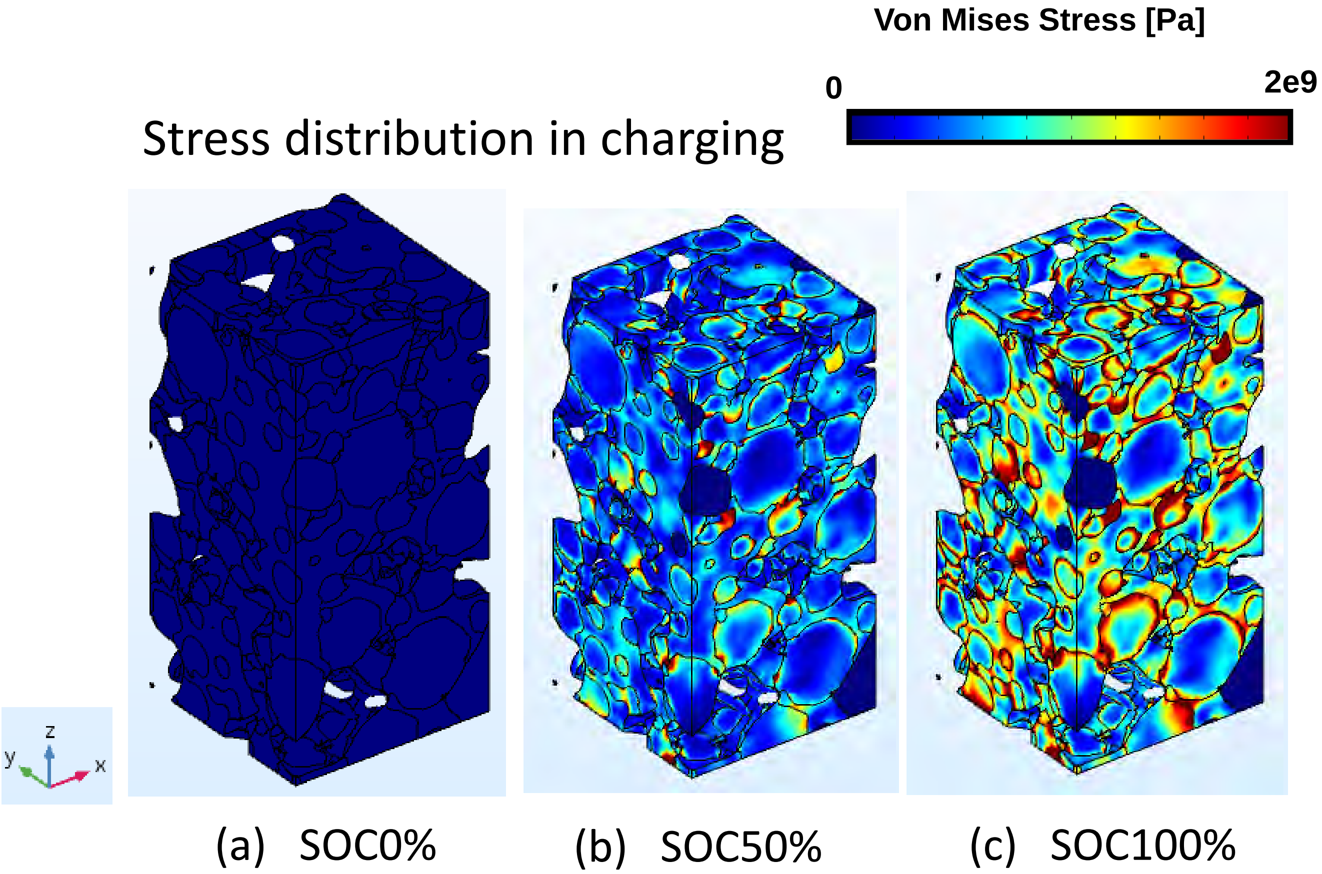
High stress on the edge of active material

Multiphysics Simulation based on 3D microstructure

Charge/discharge FEM simulation: Virtual microstructure

virtual electrode structures composed of random packed sphere particles and binder have been generated.



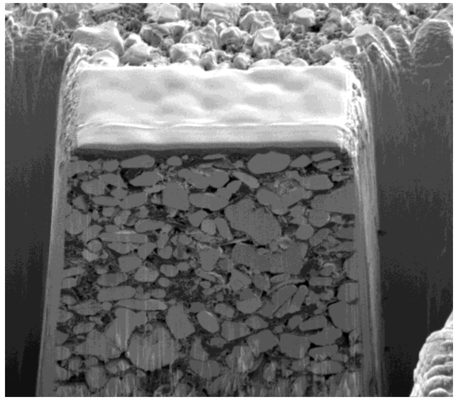


Physico-chemical Simulation using FIB-SEM image

"Quasi-3D" model with a FIB-SEM image

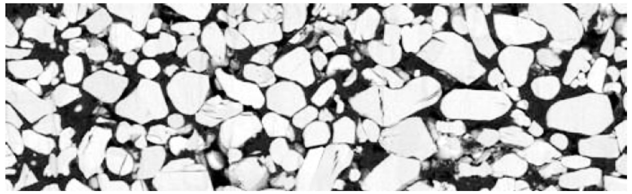
"Quasi-3D" model has been applied to a FIB-SEM image of typical positive electrode

FIB-SEM



$Li(Ni_{1/3}Mn_{1/3}Co_{1/3})O_2$

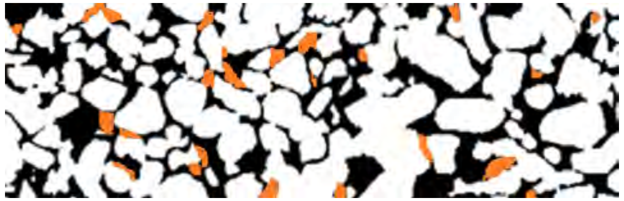
2D-SEM image



50[um]

Segmented image

432px × 125px

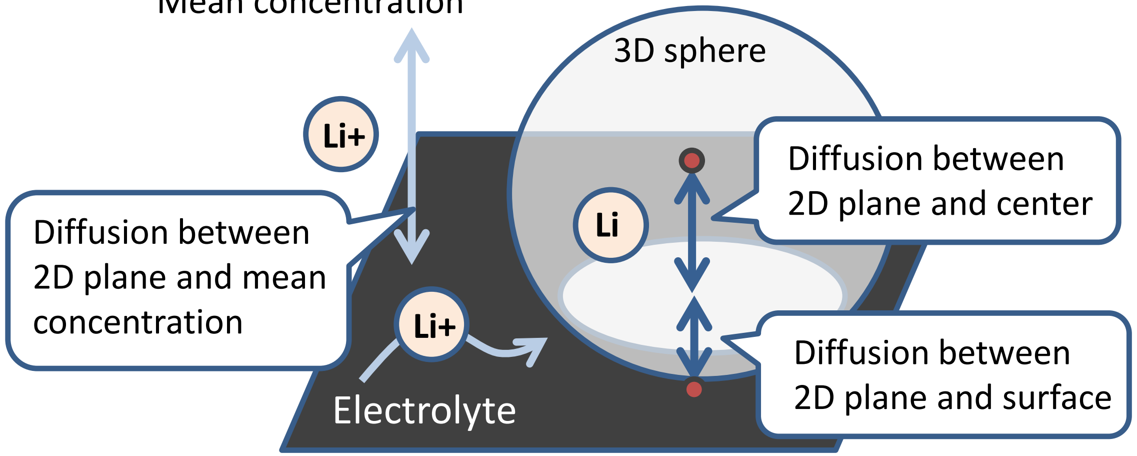


50[um]

☐ Active material

■ Electrolyte

■ Binder/Additives



Diffusion resistance

Li diffusion in solid phase : Correction term

$$\frac{\partial c_s}{\partial t} = \nabla\cdot(-D_s\nabla c_s) + \frac{q_{surf}-q_{center}}{r}$$

c_s: Li concentration
D_s: Diffusion coefficient

Electrolyte resistance

Electric potential of liquid phase :

$$\nabla\cdot\left[-\sigma_l\nabla\phi_l + \frac{2\sigma_l RT}{F}(1-t_+)\nabla \ln c_l\right] = i_{tot}$$

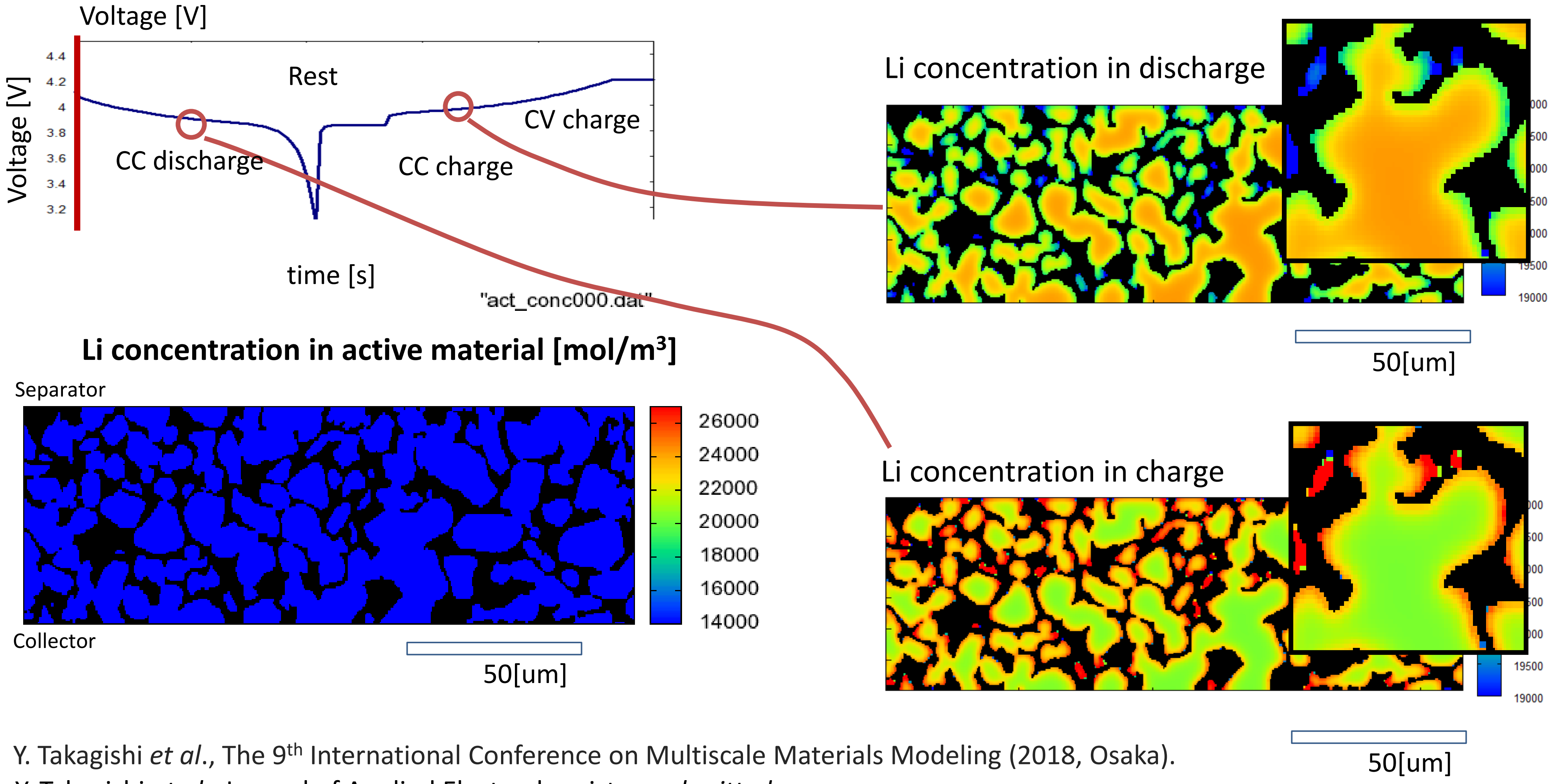
Mass balance : Correction term

$$\frac{\partial c_l}{\partial t} = \nabla\cdot(-D_l\nabla c_l) + D_L\frac{\bar{c}_z - c_L}{l^2}$$

Y. Takagishi et al., The 9th International Conference on Multiscale Materials Modeling (2018).

Physico-chemical Simulation using FIB-SEM image

Concentration of Li in active material



Y. Takagishi *et al*., The 9th International Conference on Multiscale Materials Modeling (2018, Osaka).
Y. Takagishi *et al*., Journal of Applied Electrochemistry, *submitted*.

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Topics

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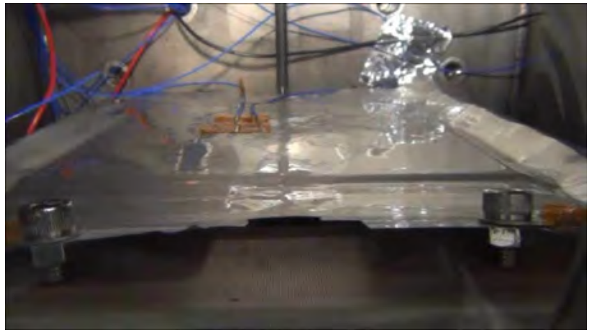
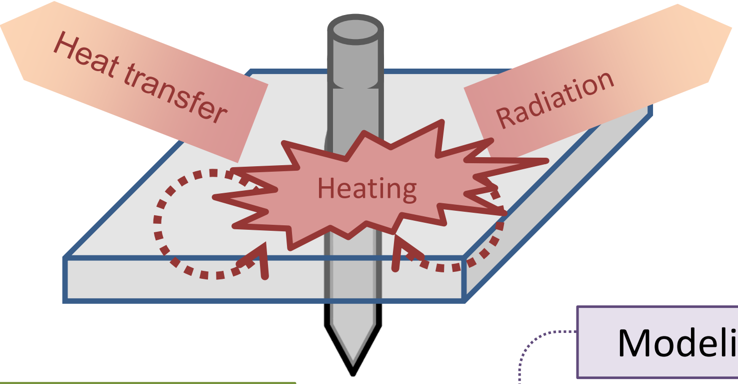
Battery Safety Simulations, Nail penetration test

Nail penetration test





Electrochemical reaction heating

Joule heating $Q_{Joule} = i_j \nabla \phi_j$

Reaction heating $Q_{reac} = i\left(\phi_s - \phi_l - E_{eq} + T\frac{\partial E_{eq}}{\partial T}\right)$

Physico-chemical model

Electrode potential:
Poisson Equation

$$\mathbf{i}_s = -\sigma_s \nabla \phi_s$$

Electrolyte potential :
Nernst-Plank Equation

$$\frac{\partial c_s}{\partial t} = \nabla \cdot (-D_s \nabla c_s)$$

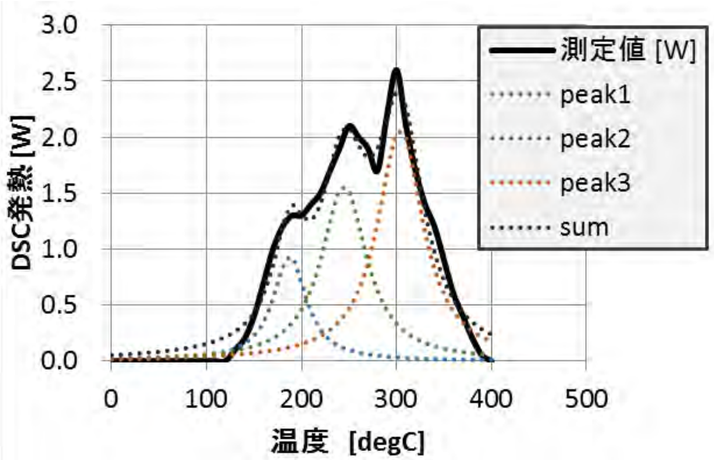
Li conc. in AP

$$i_{loc} = i_0\left(\exp\left(\frac{\alpha_a F \eta}{RT}\right) - \exp\left(\frac{-\alpha_c F \eta}{RT}\right)\right)$$

Electrochemical reaction:
Butler-Volmer Equation

$$\nabla \cdot \left(-\sigma_l \nabla \phi_l + \frac{2\sigma_l RT}{F}\left(1 + \frac{\partial \ln f}{\partial \ln c_l}\right)(1 - t_+)\nabla \ln c_l\right) = i_{tot}$$

Modeling of thermal decomposition based on DSC test

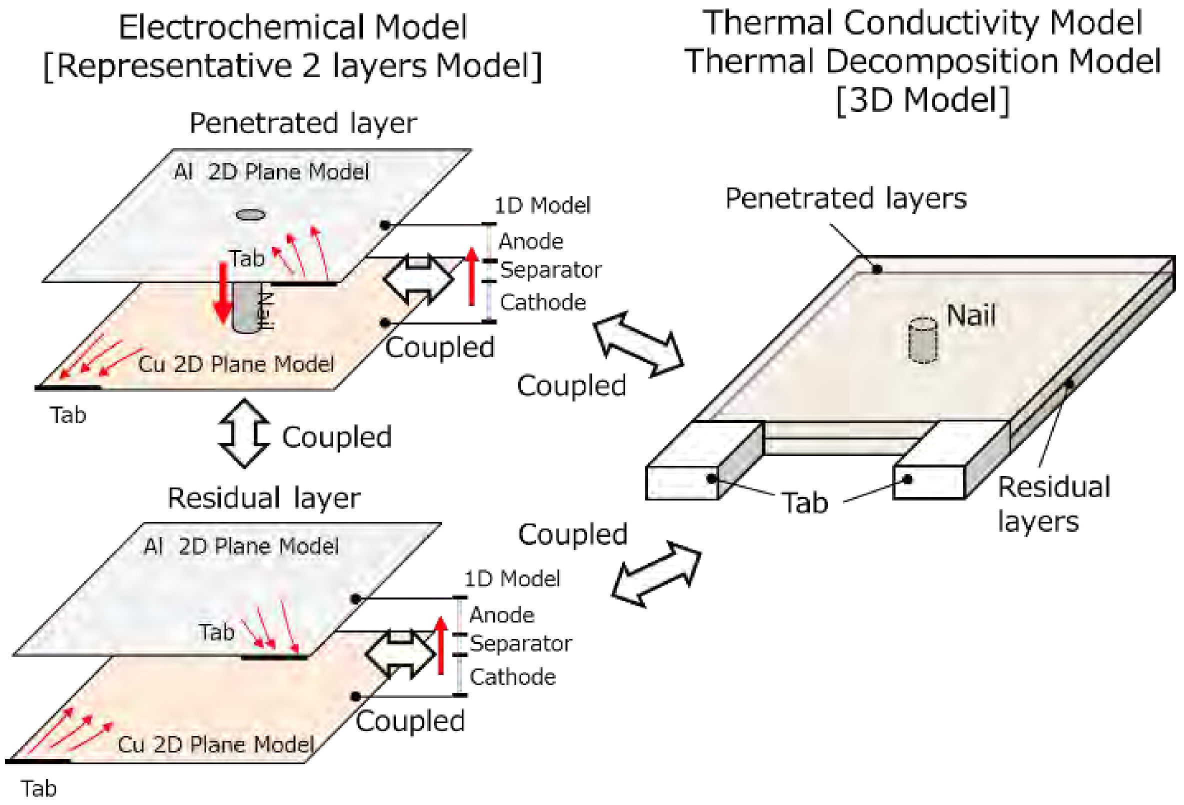


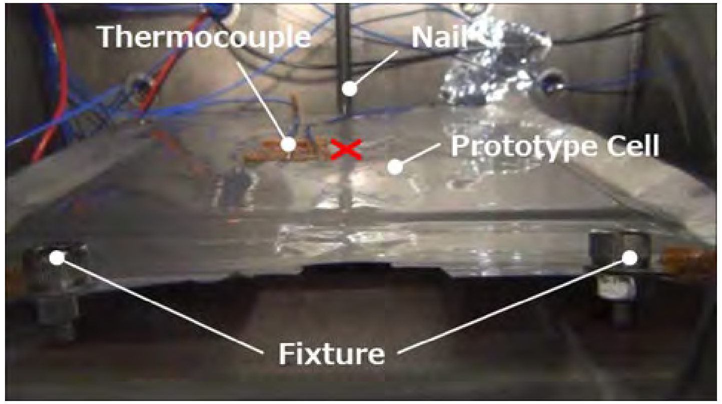
$$k = \gamma exp\left(-\frac{E_a}{RT}\right)x^n(1-x)^m(-lnx)^p$$

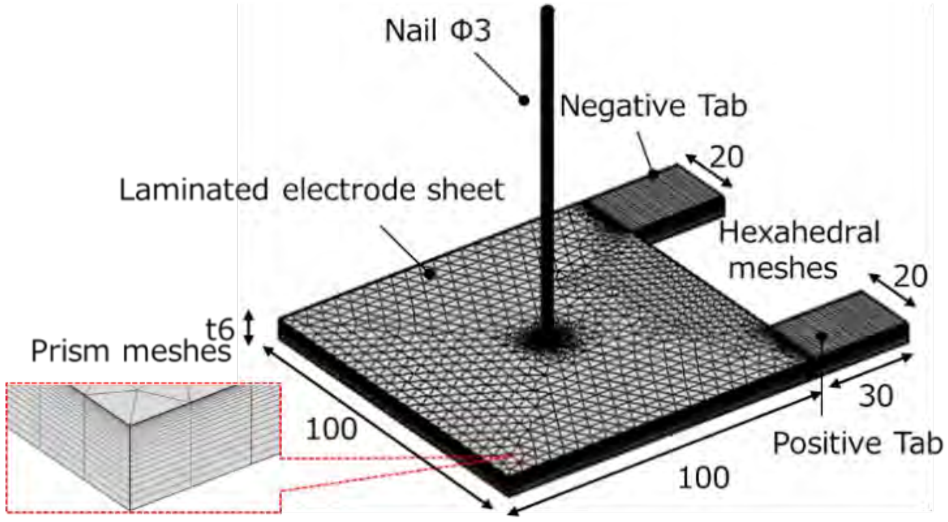
Battery Safety Simulations, Nail penetration test

"Tri-bred model"

The 1D-2D-3D coupled model taking into account of migration of the nail and thermal decomposition reaction.





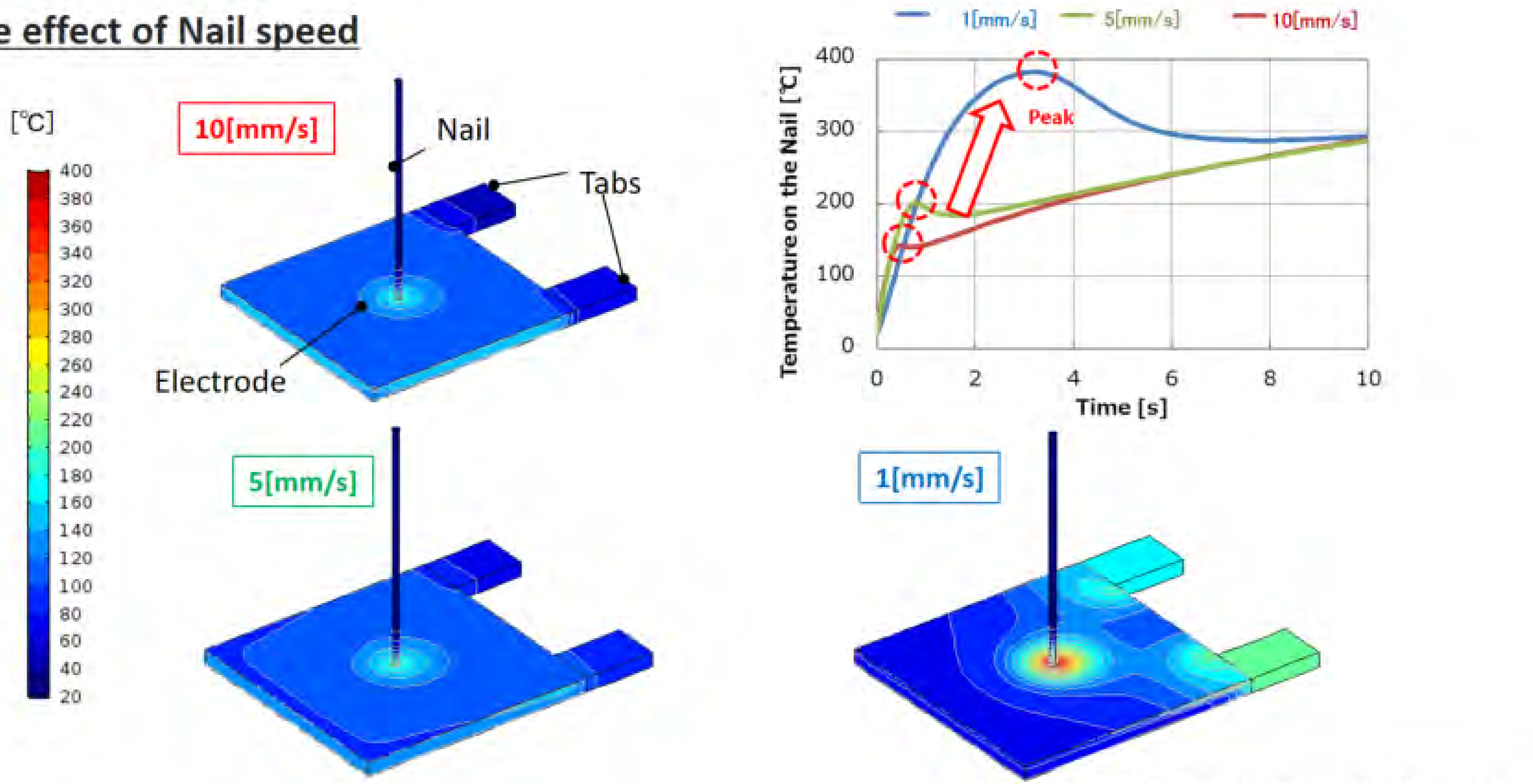


T. Yamanaka *et al*., Journal of Power Sources, 416:132-140.

Battery Safety Simulations, Nail penetration test

"Tri-bred model"

The effect of Nail speed

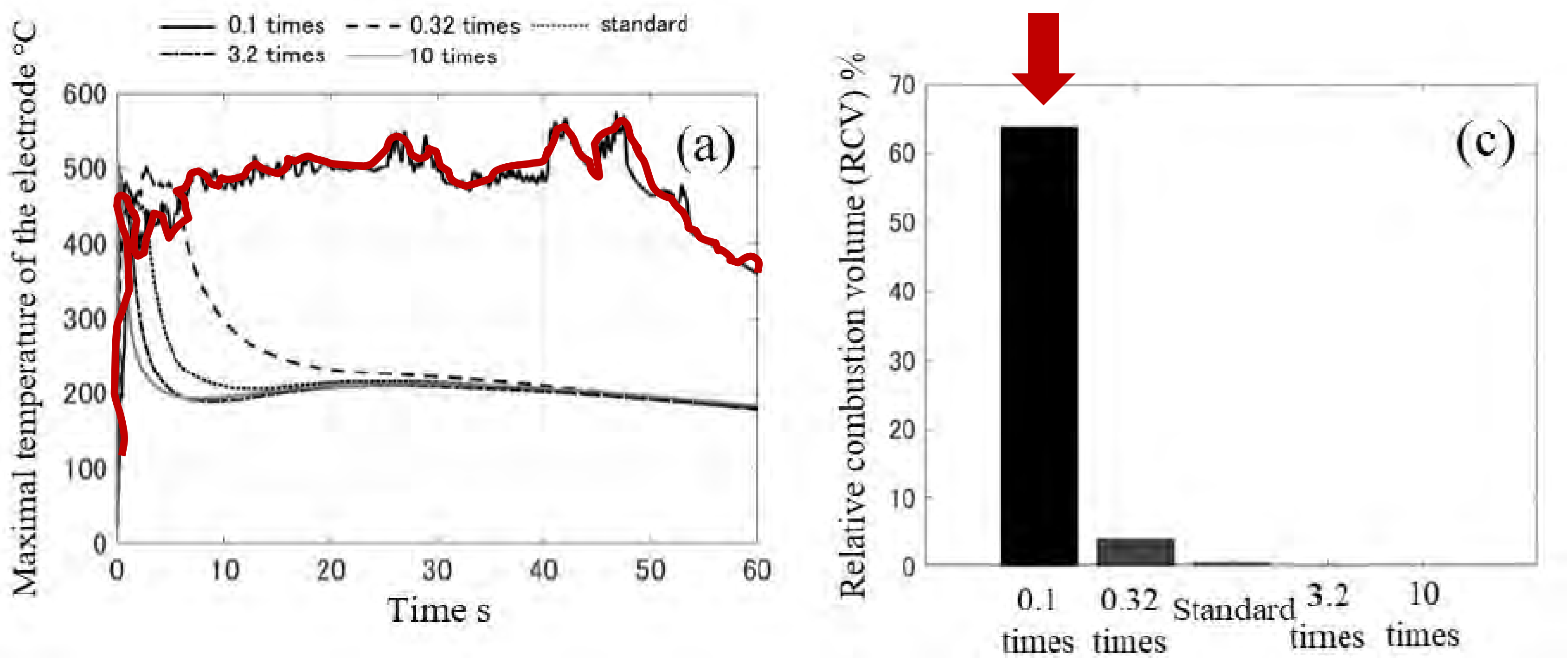


T. Yamanaka *et al.*, Journal of Power Sources, 416:132-140.

Battery Safety Simulations, Nail penetration test

"Tri-bred model"

The effect of Nail position

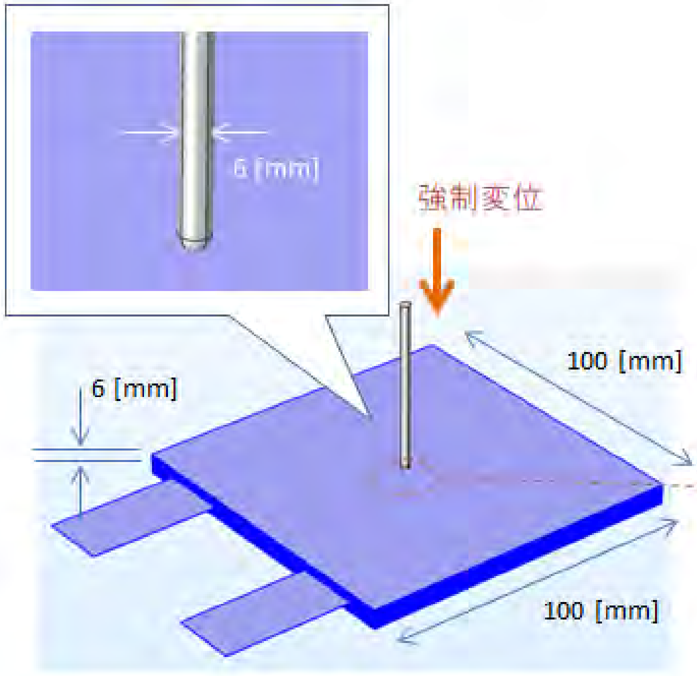


When the nail speed 0.1[mm/s], RCV is 100 times higher than those of standard condition,
And the total time spent in excess of 300 ℃ is longer.
The maximal temperature is 83 ℃ higher than the standard condition.

Battery Safety Simulations, Nail penetration test

Multi-scale modeling of indentation test

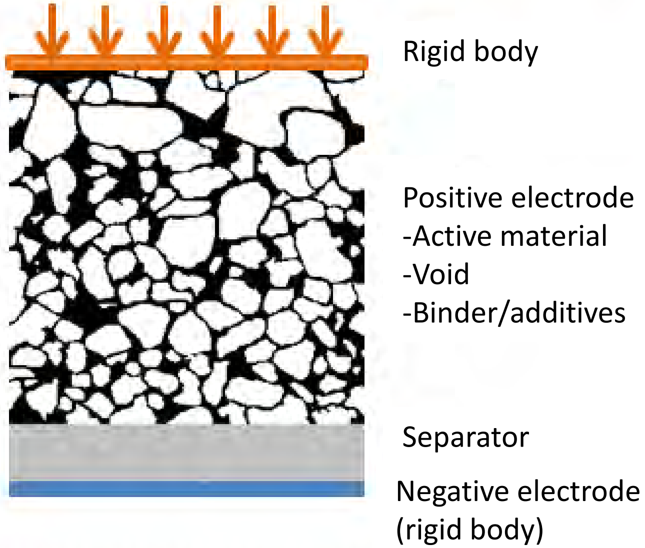
Macro-scale simulation
(Stress simulation using FEM)



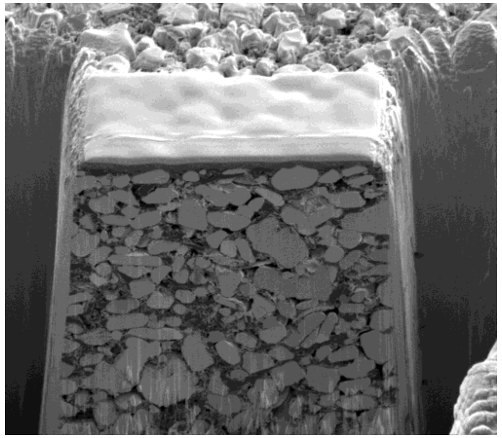
変位履歴

Micro-scale simulation
(Deformation using voxel)

- Spring-dumper model
- Current conservation
- Energy balance
- No phase change, no gas generation



FIB-SEM

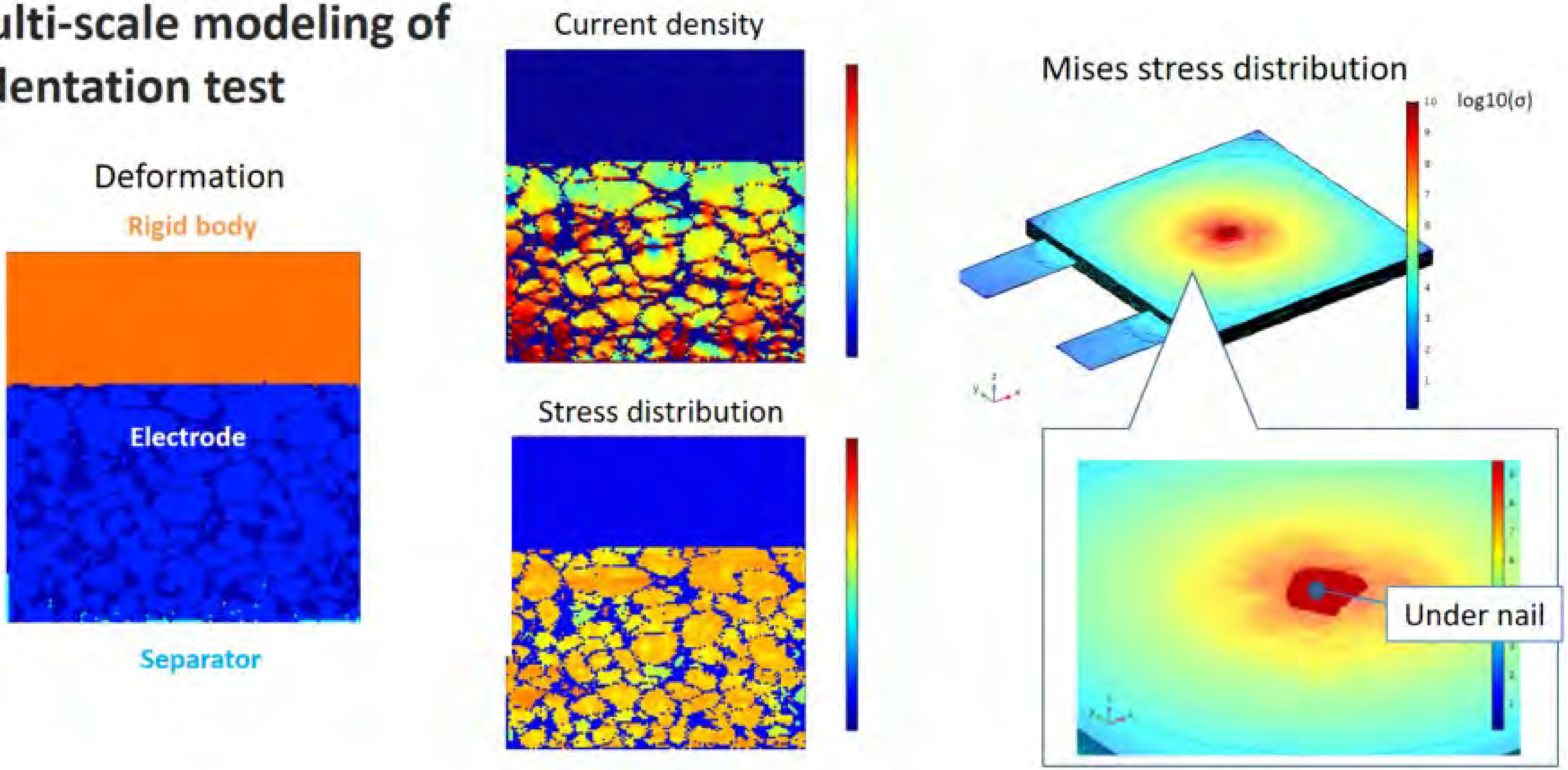


$Li(Ni_{1/3}Mn_{1/3}Co_{1/3})O_2$

Y. Takagishi *et al.*, Battery Symposium Japan (2017).

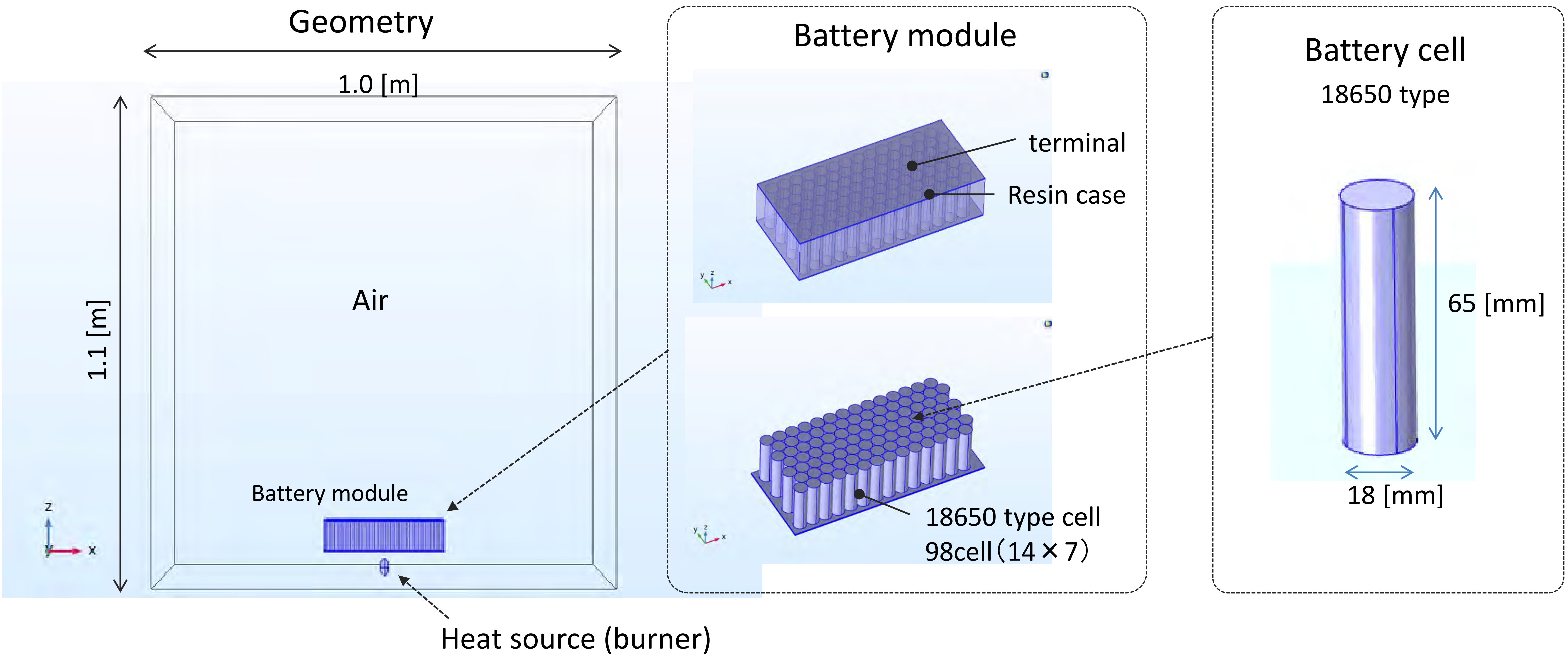
Battery Safety Simulations, Nail penetration test

Multi-scale modeling of indentation test



Y. Takagishi *et al.*, Battery Symposium Japan (2017).

Burning test of 18650 module

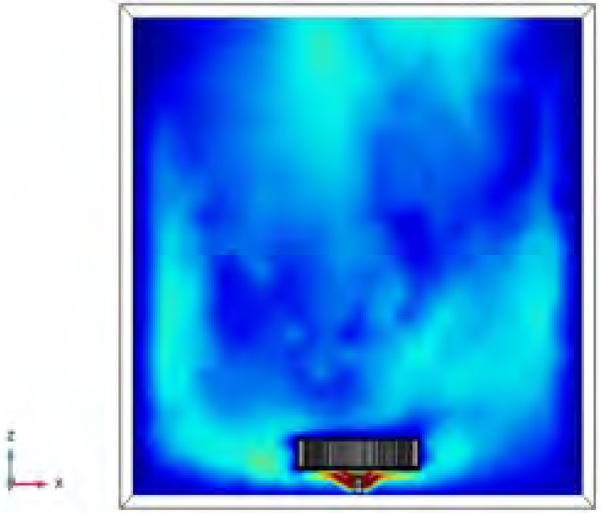


Y. Takagishi *et al.*, Battery Symposium Japan (2020).

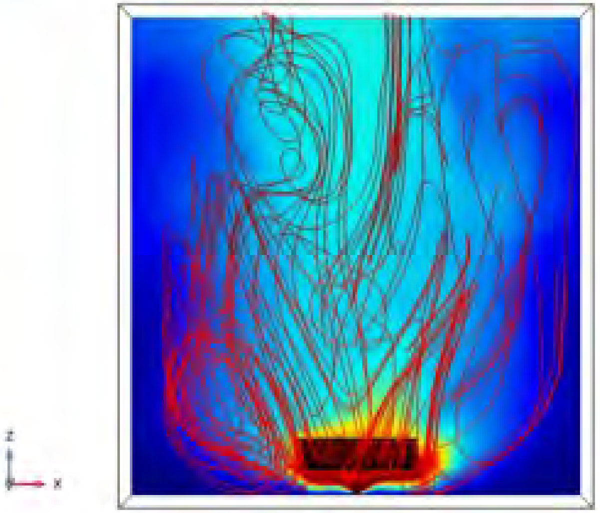
Battery Safety Simulations, Burning test

Burning test of 18650 module

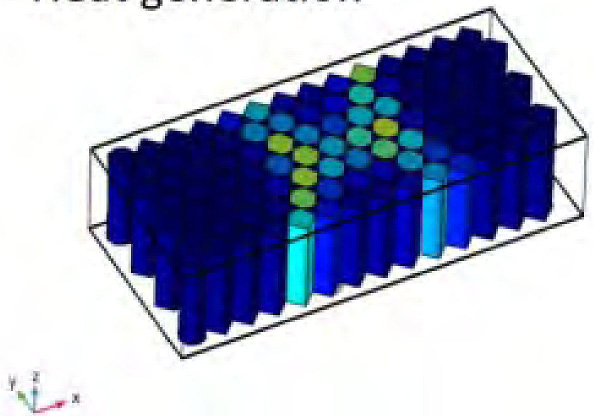
Gas flow

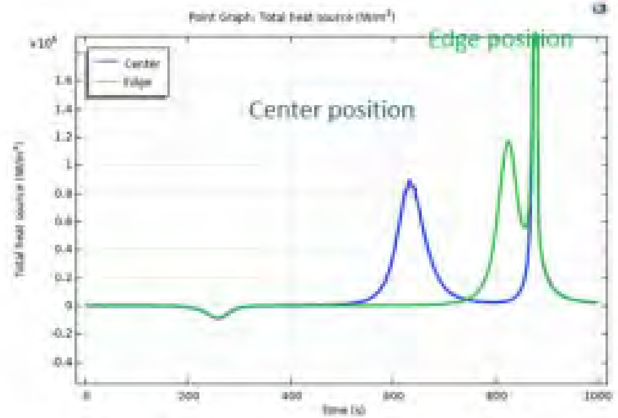


Temperature



Heat generation





Y. Takagishi *et al.*, Battery Symposium Japan (2020).

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Topics

- ☐ Equivalent Circuit Model for Battery Management System
- ☐ Physico-chemical Simulation using FIB-SEM image
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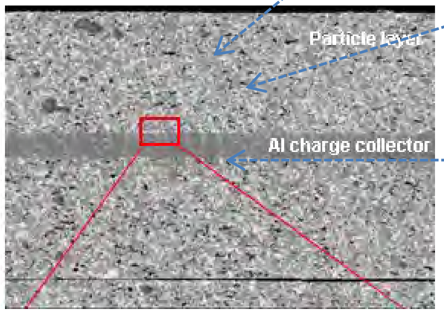
- ☐ Machine Learning, Deep Learning

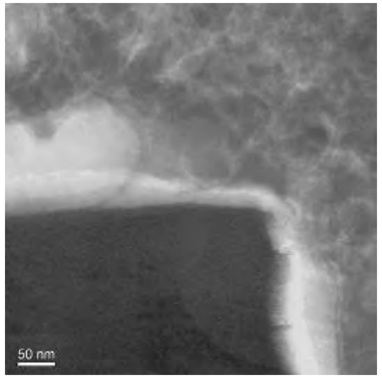
Battery Degradation Simulation

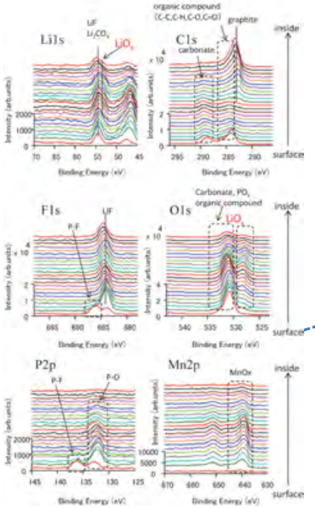
Degradation mechanism of Li-ion battery

電極合剤



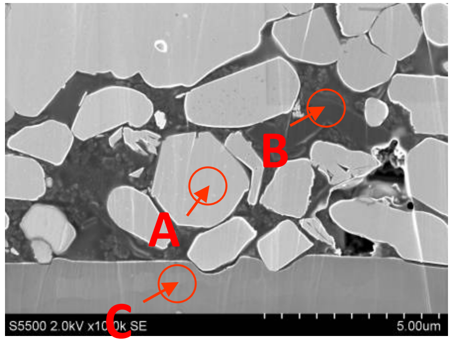
SEI thin layer growth on graphite particles



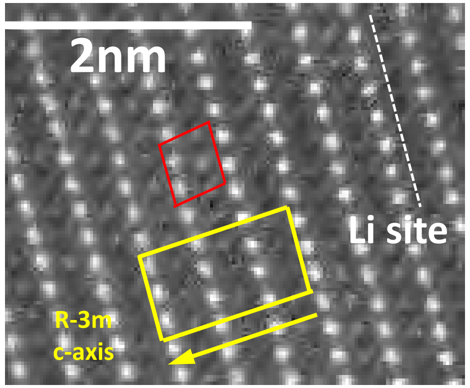


Two kinds of SEI (organic / inorganic)

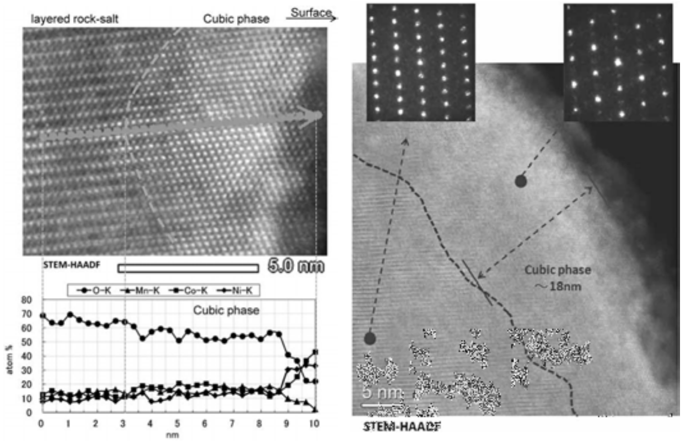
Electrode exfoliation



Phase transition on the surface of NMC particles



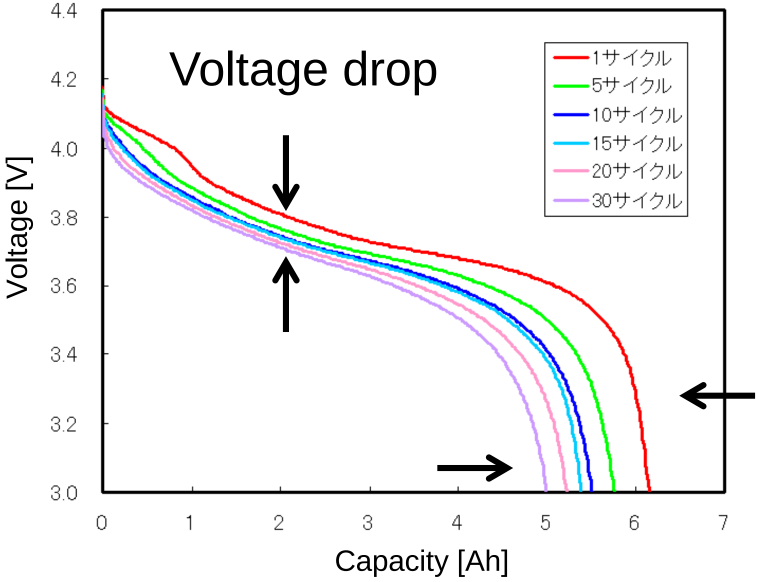
SEGI *et al.*, Battery Symposium（Osaka, 2013）



Battery Degradation Simulation

Degradation in charge/discharge cycling

Cycle properties of $LiCoO_2/C$

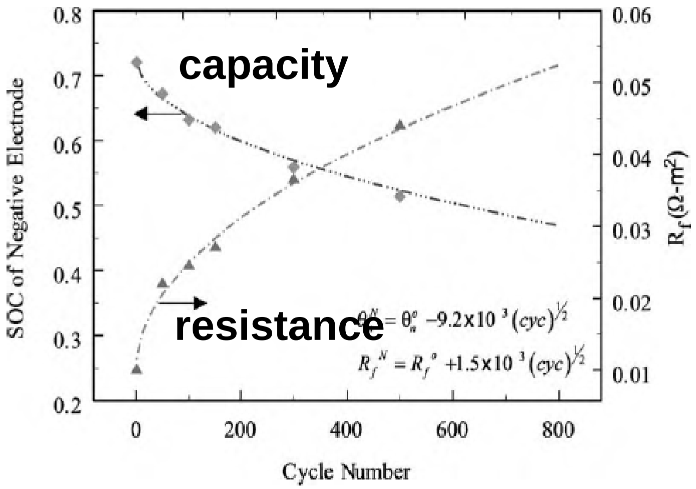


Tsubota, et.al., Battery Symposium.（Tokyo, 2011）

Empirical √t - law

resistance $R = R^0 + k_1(cycle)^{1/2}$

capacity（SOC） $\theta = \theta^0 - k_2(cycle)^{1/2}$



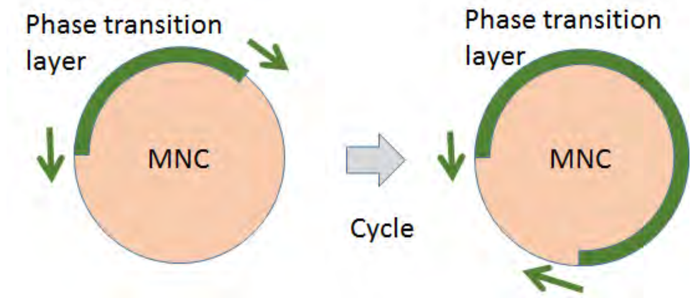
*P. Ramadass *et al*., Journal of Power Source **123** (2003) 230.

Mechanics of capacity fade and resistance increase is a blackbox

Battery Degradation Simulation

Physics-based degradation model

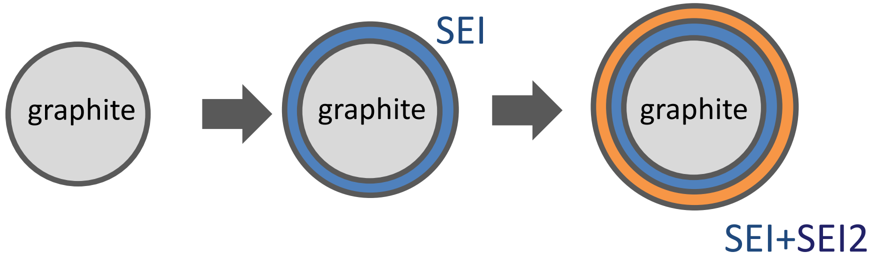
Growth of cubic layer on positive AP



Coverage ϑ_p :

$$\dot{\theta}_p = k(1-\theta_p)$$

Growth of two kinds of SEI on AP

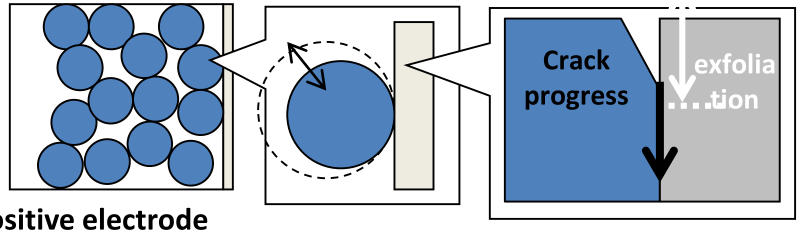


Growth rate dδ/dt : Tafel model

$$\dot{\delta} = \frac{M\rho}{F} J_s \qquad J_s = i_{0S}\, a\, 0.5F\eta_s / RT$$

$$\eta_s = \eta_{s,0} - R_{sei} J$$

Electrode exfoliation



positive electrode

Crack progress model : Paris-law

$$\frac{da}{dn} = C_0 \Delta K^m$$

$$R = R_0 \frac{a_0}{a_0 - a}$$

The range of fluctuation of a stress intensity factor

$$\Delta K = Y \Delta S \sqrt{\pi a}$$

Y: material constant

ΔS: stress fluctuation range

**Capacity is not decreased in this process.

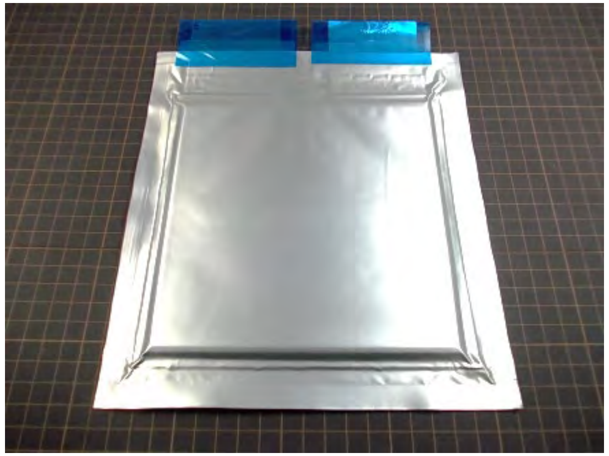
Battery Degradation Simulation

Methodology: Experimental

For comparison with simulation, we have also performed charge-discharge cycling test.

Test battery（400[mAh]）

- **Positive electrode: NMC ($LiNi_{1/3}Mn_{1/3}Co_{1/3}O_2$)**
- **Negative electrode: Graphite**
- **Electrolyte: 1M $LiPF_6$ / EC:DEC=1:1**
- **Thickness:**
  - **Positive electrode 50 [um]**
  - **Negative electrode 50 [um]**
  - **Separator 25 [um]**



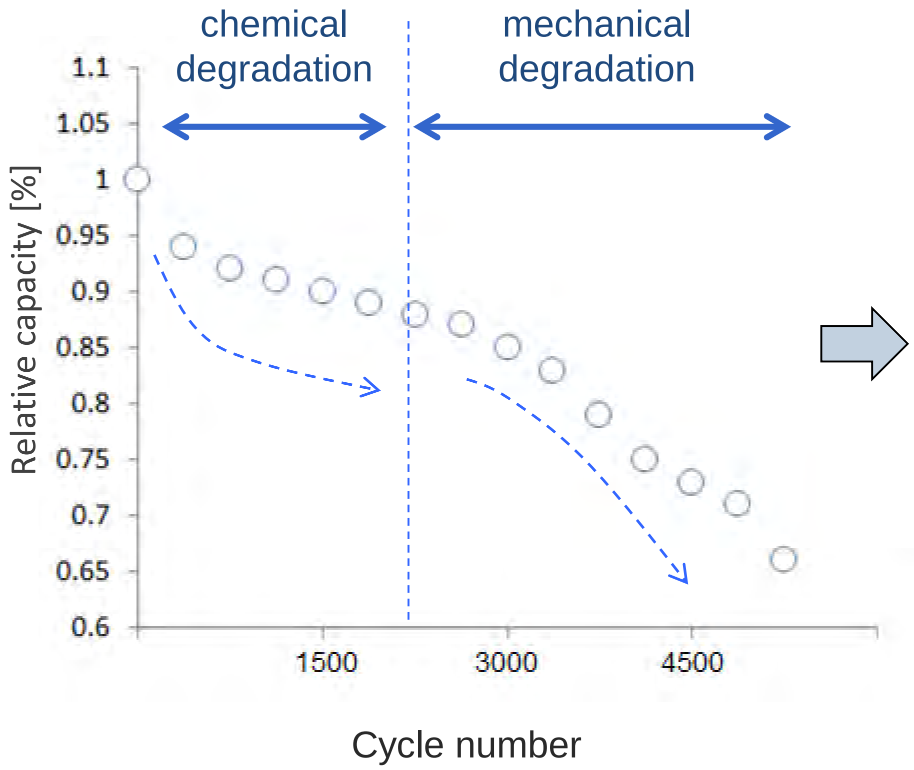
Cycle conditions

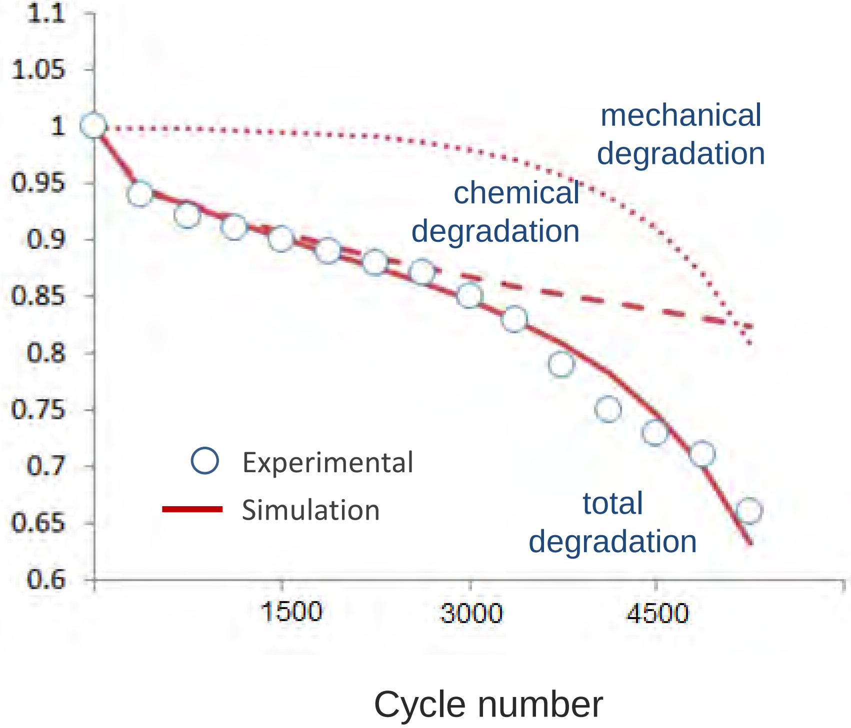
- **Charge/discharge rate: 2C (CC-CV)**
- **Rest: 10 [min]**
- **Maximum cycle number: 6100**
- **Voltage range: 4.2 [V] - 2.7 [V]**
- **Temperature: 25 [degC]**

Battery Degradation Simulation

Physics-based degradation model

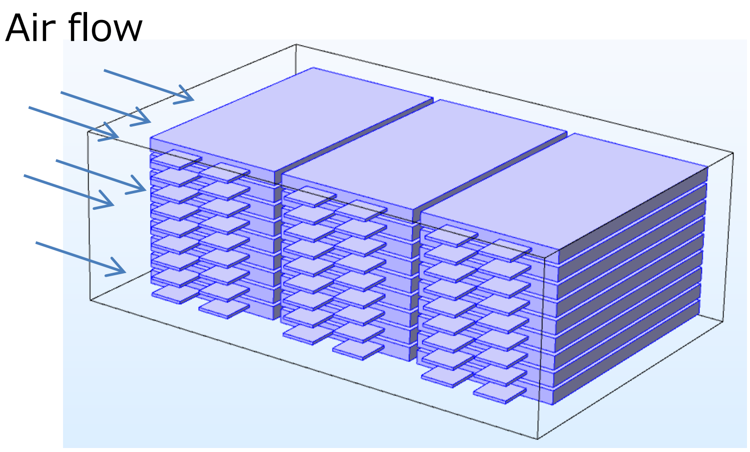
Comparison of capacity degradation between simulation and experiment



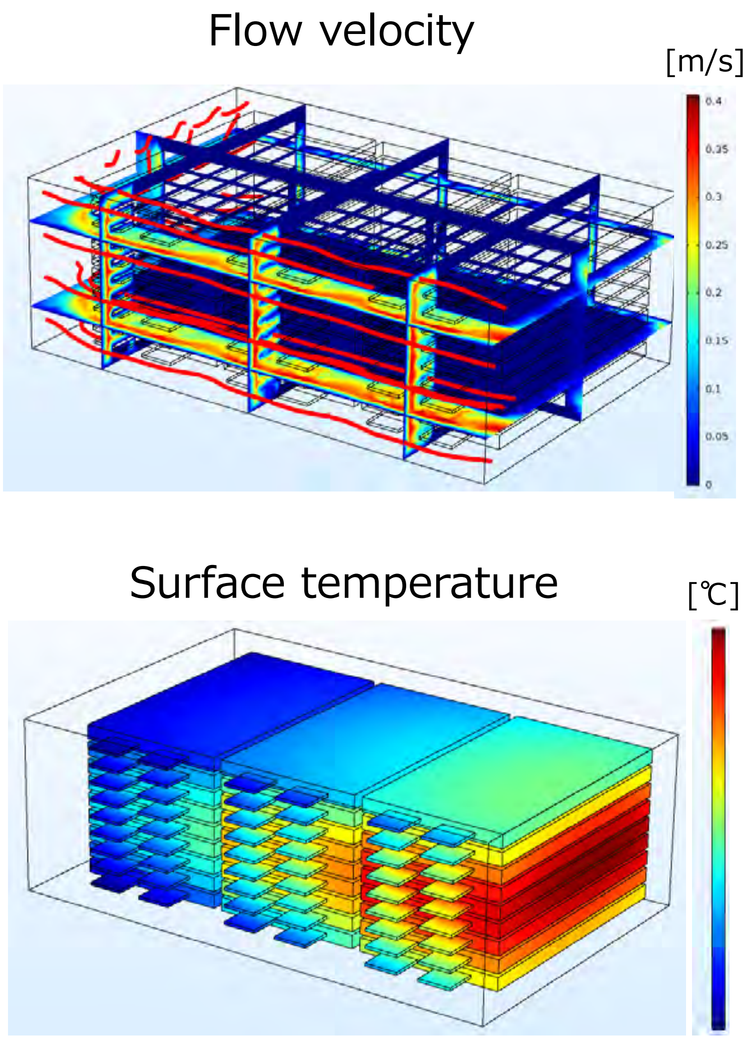


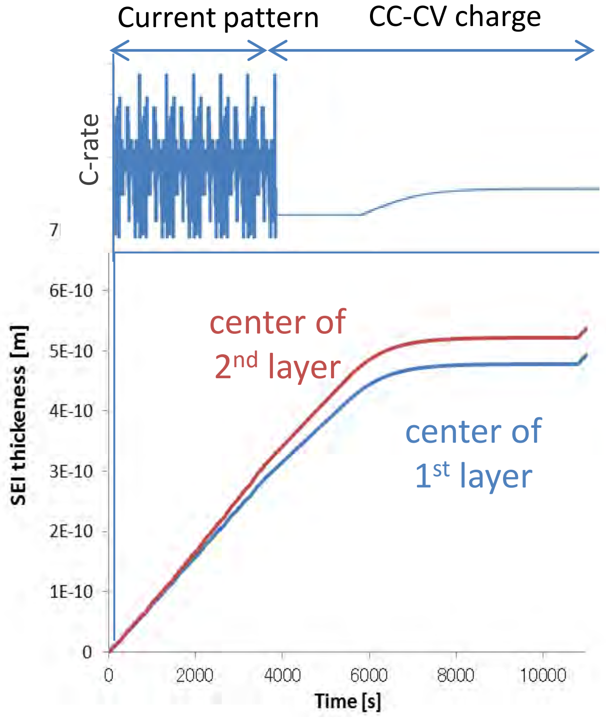
Y. Takagishi *et al*., Batteries Event 2015 (Nice, France).

Battery module degradation



Laminated test battery（electrode: 50[cm2]）
9S3P module with air cooling





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Topics

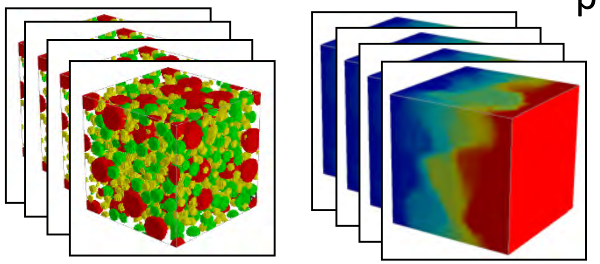
- ☐ Equivalent Circuit Model for Battery Management System
- ☐ Physico-chemical Simulation using FIB-SEM image
- ☐ Battery Safety Simulations, Nail penetration test, Burning test
- ☐ Battery Degradation Simulation
- ☐ Machine Learning, Deep Learning

Machine Learning with CAE

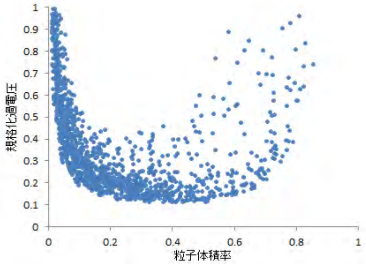
Machine Learning for Designing Meso-scale Structure of Porous electrode

3-D simulation based on voxel

Generate structures



Thousands of high throughput physico-chemical simulation



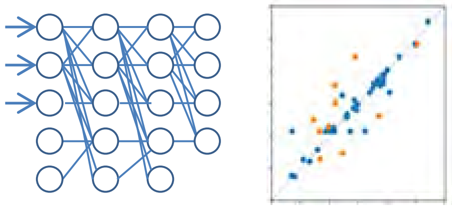
Condition parameters (predictor)

Particle volume ratio
Particle radius
Contact area between particles
Conductivity of matrix
Diffusivity of Li in particles

Databse

Regression

Artificial Neural Network

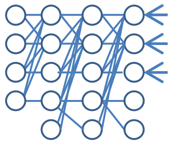


Electrode performance （Objective function）

Conductivity in matrix
Charge/discharge resistance
Capacity density, Durability

Inverse problem

<u>Artificial Neural Network</u>
(simple method)



<u>Bayesian process</u>

$$Y = \hat{\Phi} + \varepsilon$$

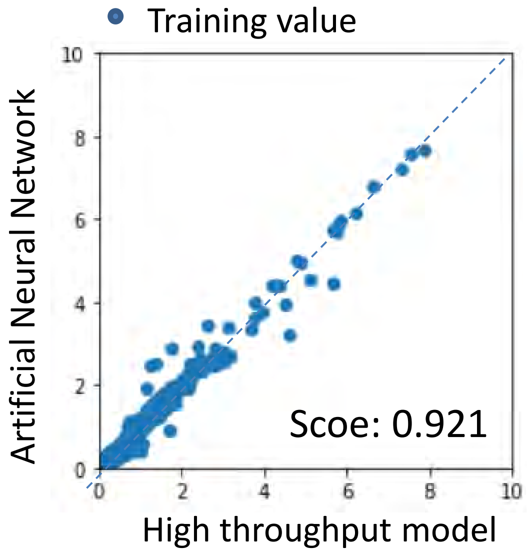
Y. Takagishi *et al*., Batteries 2019, 5, 54.

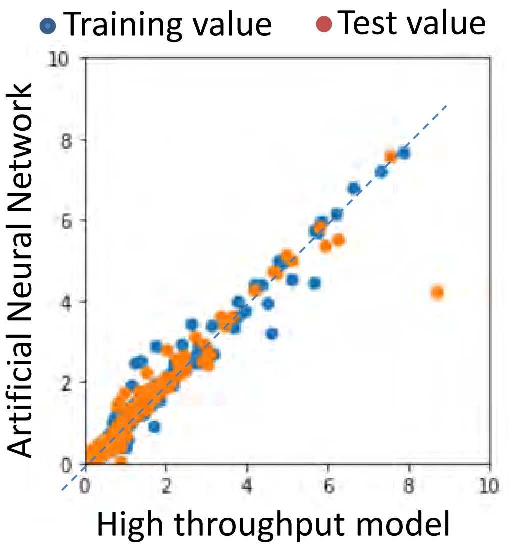
Machine Learning with CAE

Machine Learning for Designing Meso-scale Structure of Porous electrode

Artificial Neural Network regression was employed for total resistance of electrode using condition parameters.

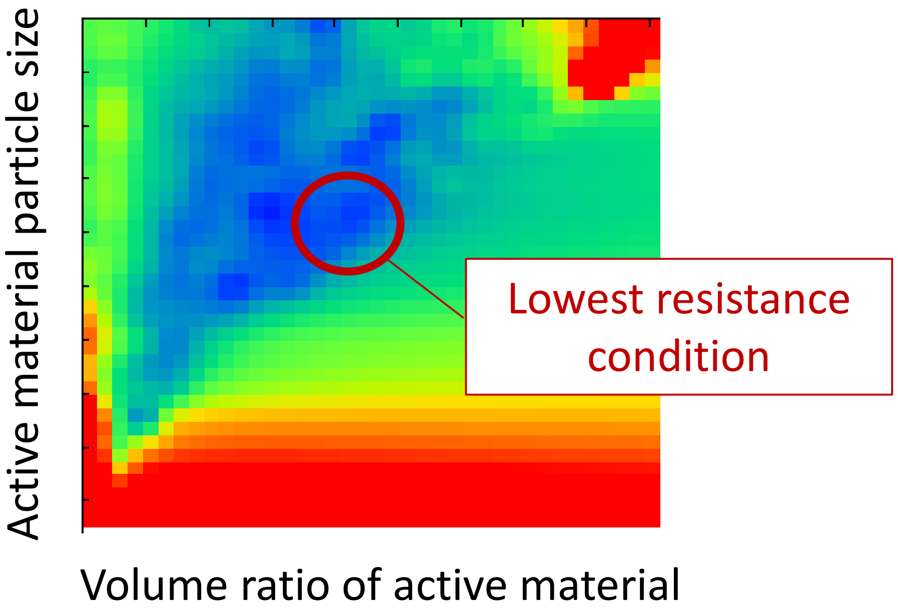
Regression of total resistance





Number of hidden layers: 3   Number of neurons: 20 in each layer
Activation function: relu   Learning coefficient: 0.001

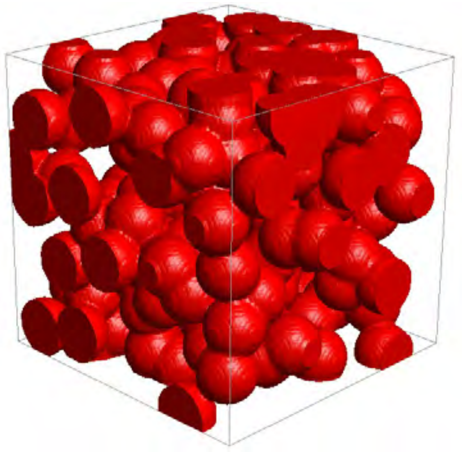
Total resistance map

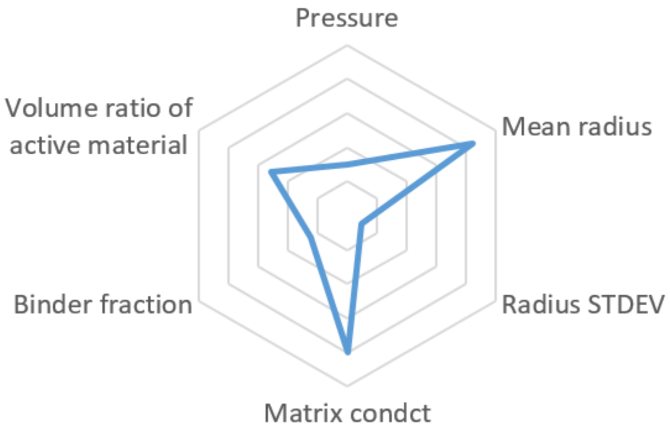


Machine Learning for Designing Meso-scale Structure of Porous electrode

The best condition parameters were determined by Bayesian optimization.

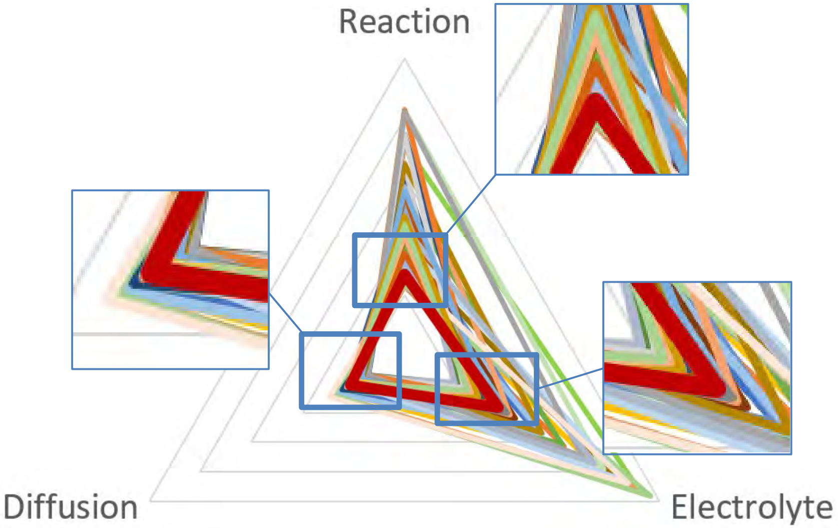
Optimum structure to reduce total resistance





Moderate particle radius and volume ratio of active material, lower pressure…

Each resistance of optimum structure



Y. Takagishi *et al.*, Batteries 2019, 5, 54.

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