早稲田大学/エイヴィエルジャパン株式会社 社会連携講座
「自動車用パワートレイン開発プロセスおよび開発手法」
シンポジウム
～Carbon Neutrality & Zero Emissions～

A Lithium-ion Cell Pseudo-Two-Dimensional Model using Real Driving Cycles

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Outline

- Background
- Previous research
- Objective
- Research method
- Experimental method
- Model construction
- Model calculation
- Conclusion

Background

Vehicles need to be electrified to address environmental issues

<Problems of electrification vehicle>

- ✓ Price
- ✓ Charging infrastructure, charging time
- ✓ Efficiency, Range

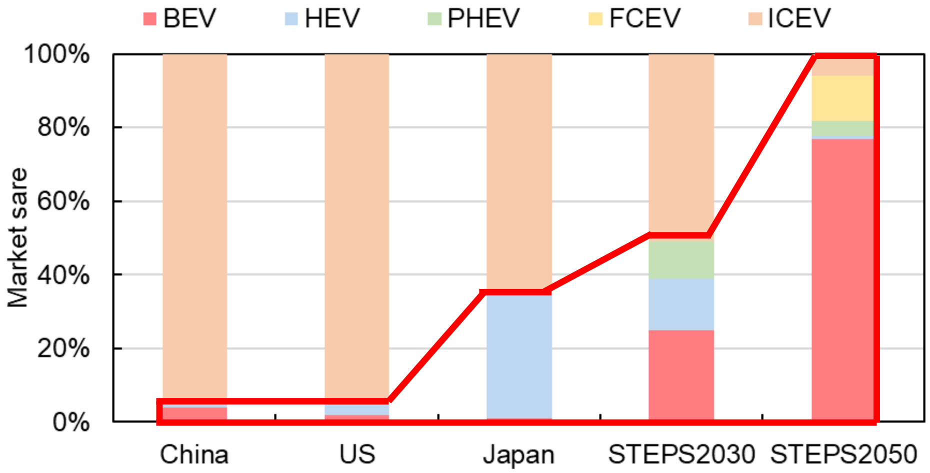
<Problems of improving efficiency>

- ✓ Heat source change
- ✓ Variety of temperature zones
- ✓ Increased complexity of thermal circuits

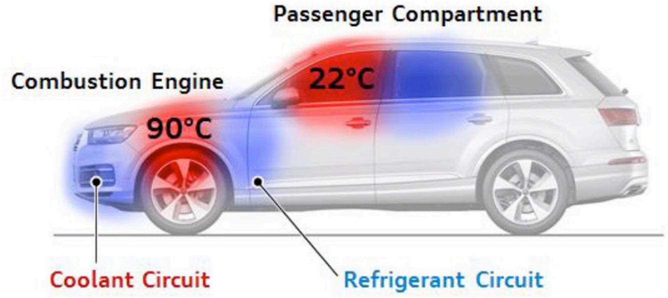
Thermal energy flow needs to be improved throughout the vehicle using MBD

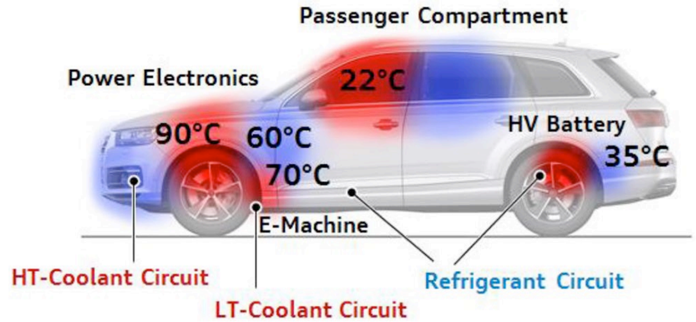
<Problems of battery modeling>

- ✓ Third parties other than OEM and battery suppliers have difficulty obtaining battery data
- ✓ Increased number of tests due to a wide range of temperatures



(1) IEA, 2021, STEPS: Stated Policies Scenario, SDS: Sustainable Development Scenario

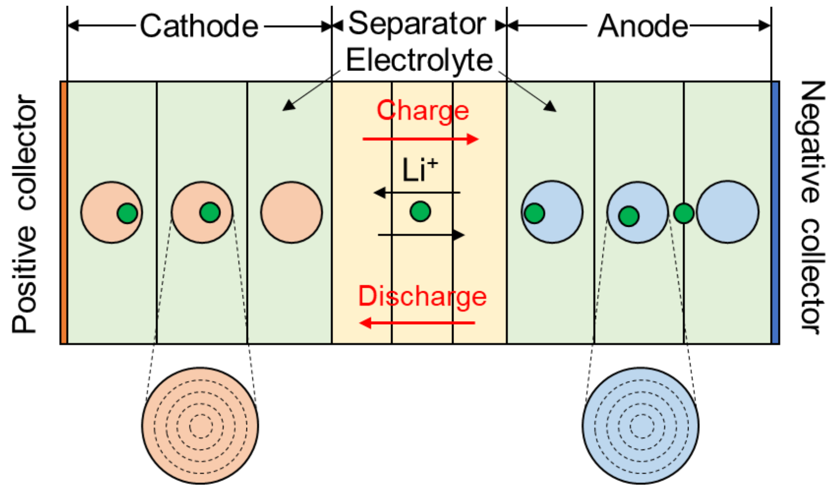




(2) Jan Christoph Menken et al., SAE international 2018

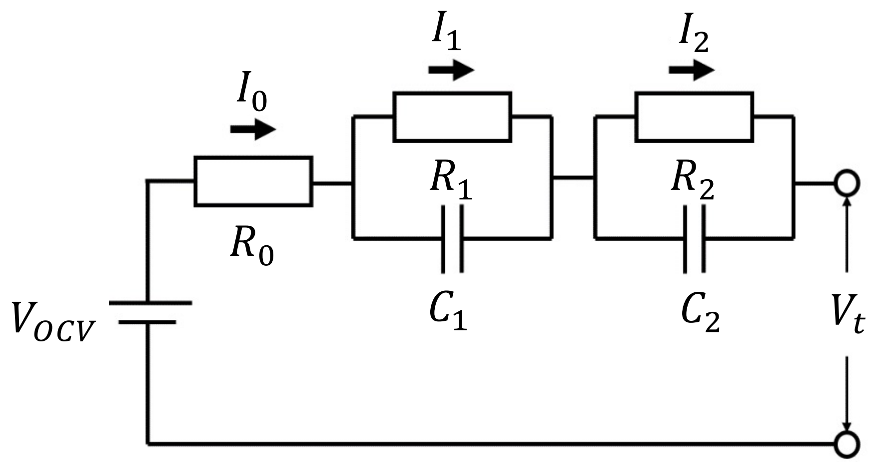
Background

P2D (Pseudo-Two-Dimensional) model



- ✓ Model lithium-ion transport phenomena and electrode reactions based on potential and lithium-ion concentration gradients

ECM(Equivalent Circuit Model)



- ✓ Model chemical phenomena with electric circuit elements

Long ⟵ Computation time ⟶ Short

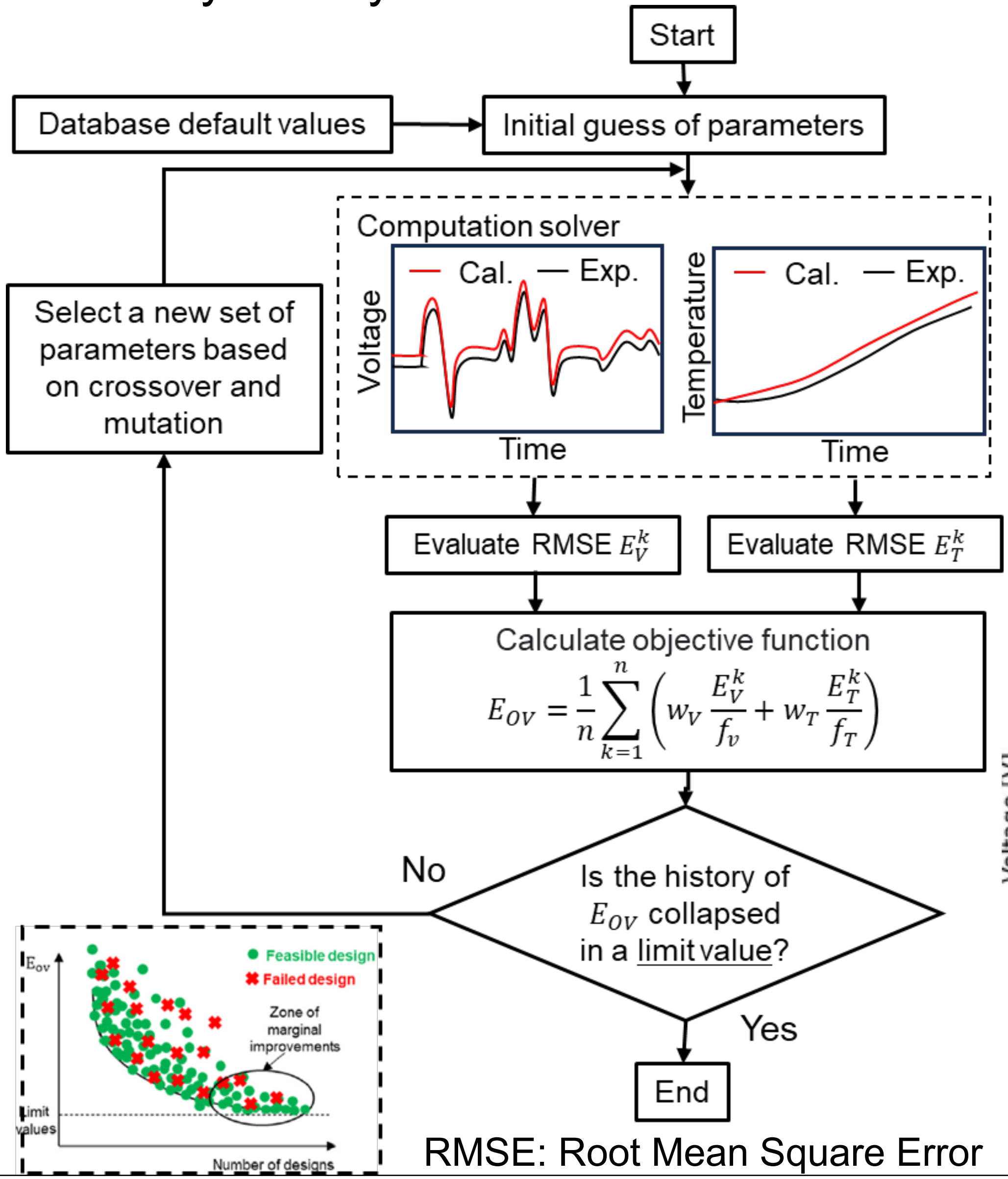
High ⟵ Predictability ⟶ Low

⇩

Fewer experiments are required

Previous research

Antonio et al. [3] atUniversidad Politécnica de Valencia have developed a method to identify battery P2D models from cell tests simulating transient driving cycles



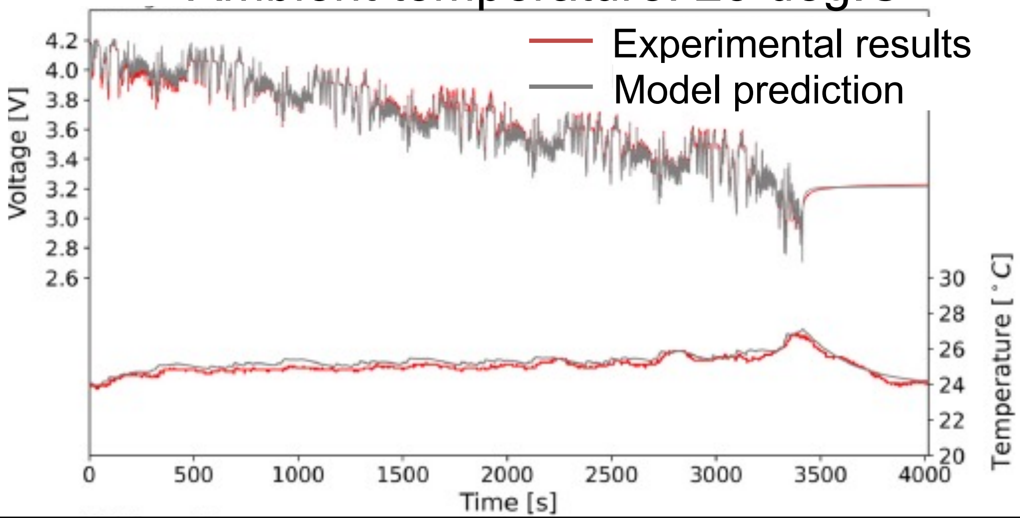
RMSE: Root Mean Square Error

Fitting parameters

Parameter	Unit	Location
Thickness	μm	Cathode, Anode, Separator
Particle size	μm	Cathode, Anode
First charge capacity	mAh/g	
First discharge capacity	mAh/g	
Constant resistance	Ω･m²	
Capacity	Ah	

Cell: LG Chem INR18650HG2(NMC 811)

Ambient temperature: 25 deg.C



Objective

<Problems of previous research and improvement>

Problems: Battery cell or experimental data of cell must be obtained

Improvement: Assuming model building from vehicle driving experiments

Modeling heat exchange between battery cells and cooling system

<Objective>

Battery P2D model construction based on vehicle driving tests

Research method

Experiment

Vehicle test

- ✓ Acquire battery and coolant data for parameter identification
- ✓ Measure

    Battery: voltage, current, SOC, temperature

    Coolant: temperature, pressure and flow rate

  during driving on the **vehicle test bench**

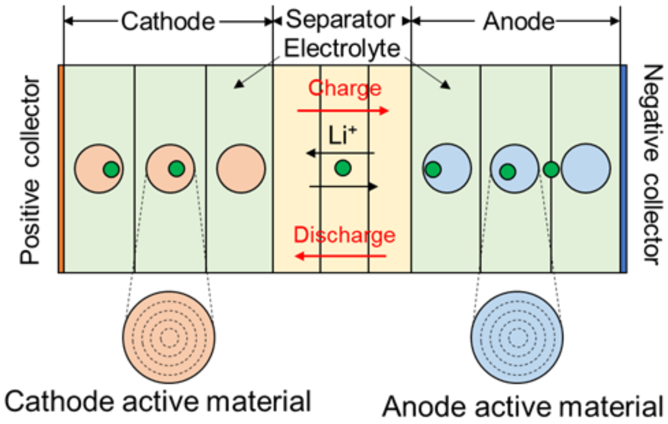
Calculation

Parameter identification

- ✓ Construct P2D model by GT-AutoLion and identify model parameters based on experimental data

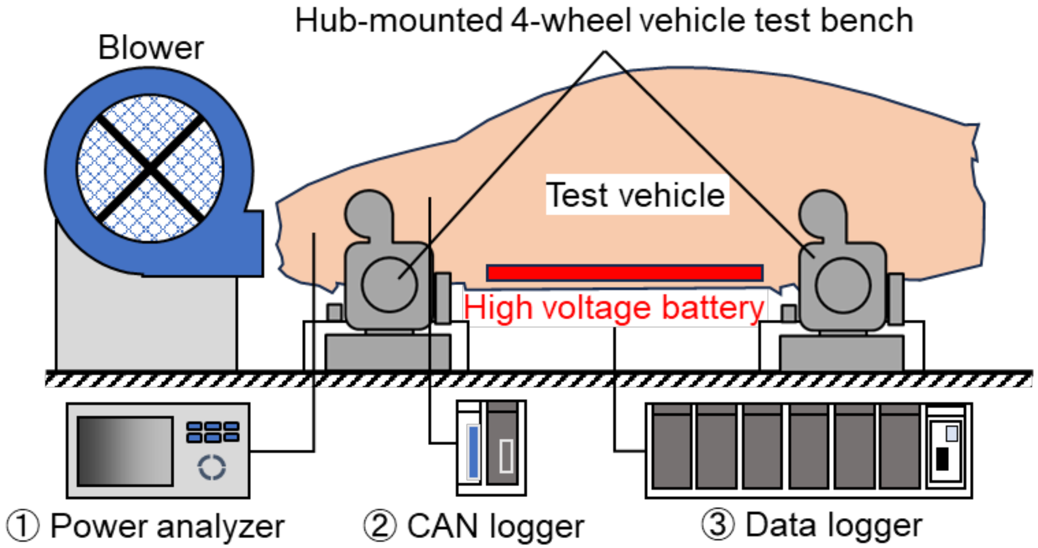
Model analysis

- ✓ Calculate the constructed model and compare calculation results and experimental results. Consider ways to improve the model



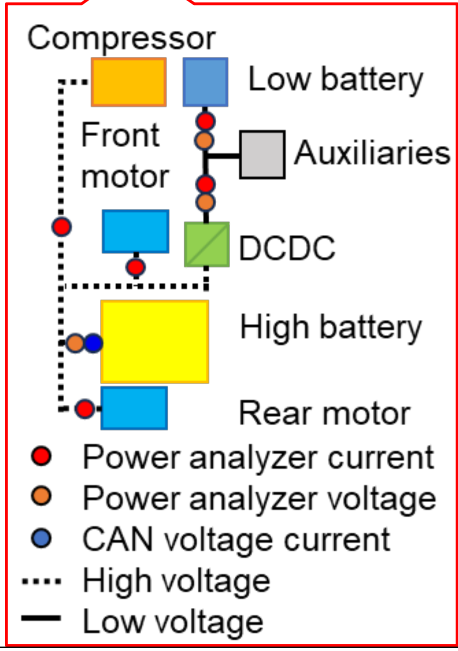
Research method

✓ Battery data was obtained by running tests on the University's vehicle test bench



Test vehicle specification[4]

Curb mass	2003 kg
Battery pack capacity	74 kWh
Coolant	Ethylene glycol 50 %
Front motor	AC Induction motor
Rear motor	AC Permanent magnet synchronous motor
Range	450 km

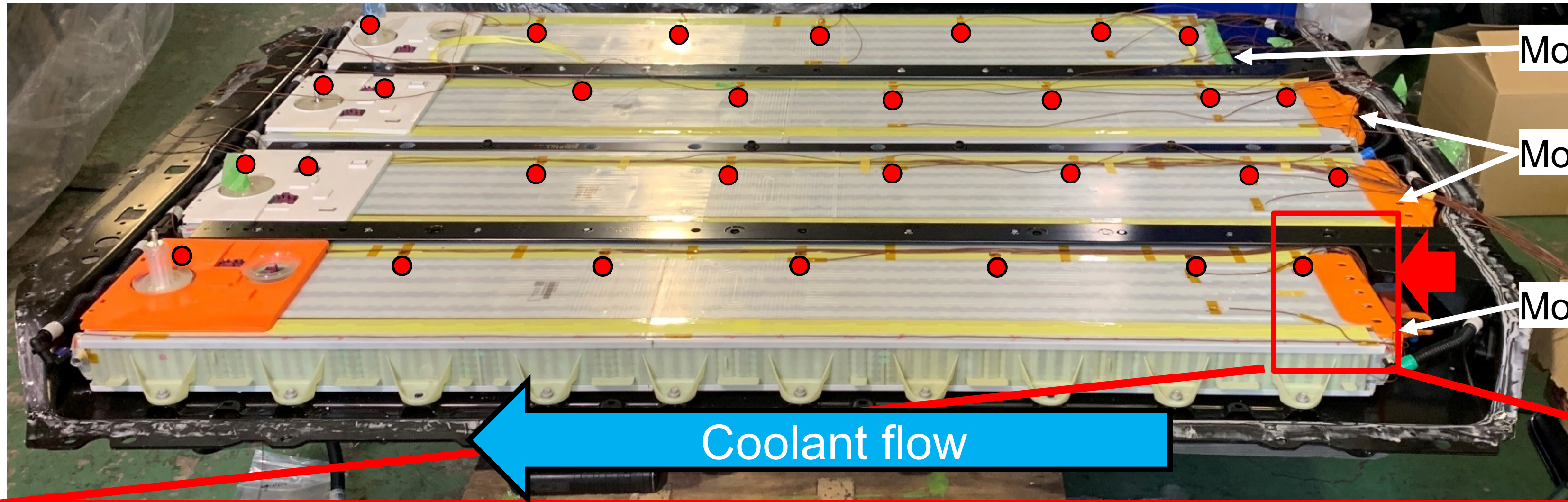


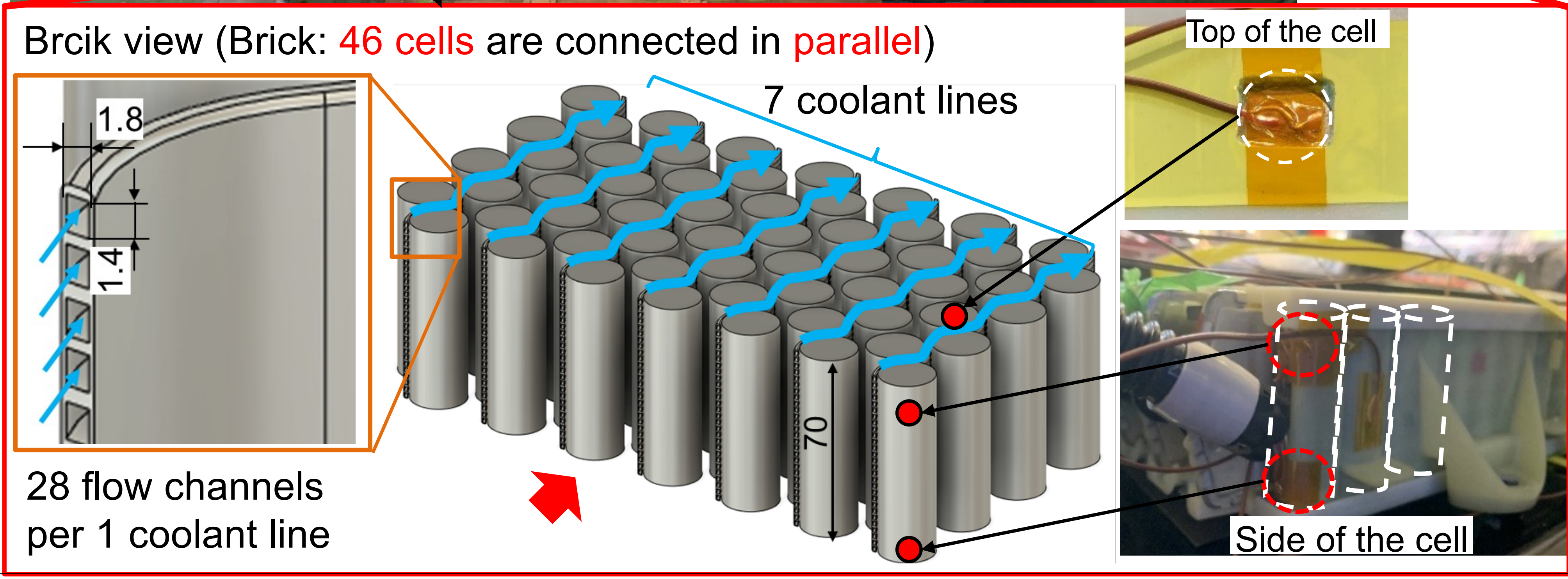
Main measurement items

Battery	Voltage
	Current
	SOC (State of Charge)
	Temperature
Motor	Torque
	Speed
Driver	Accelerator position

Research method

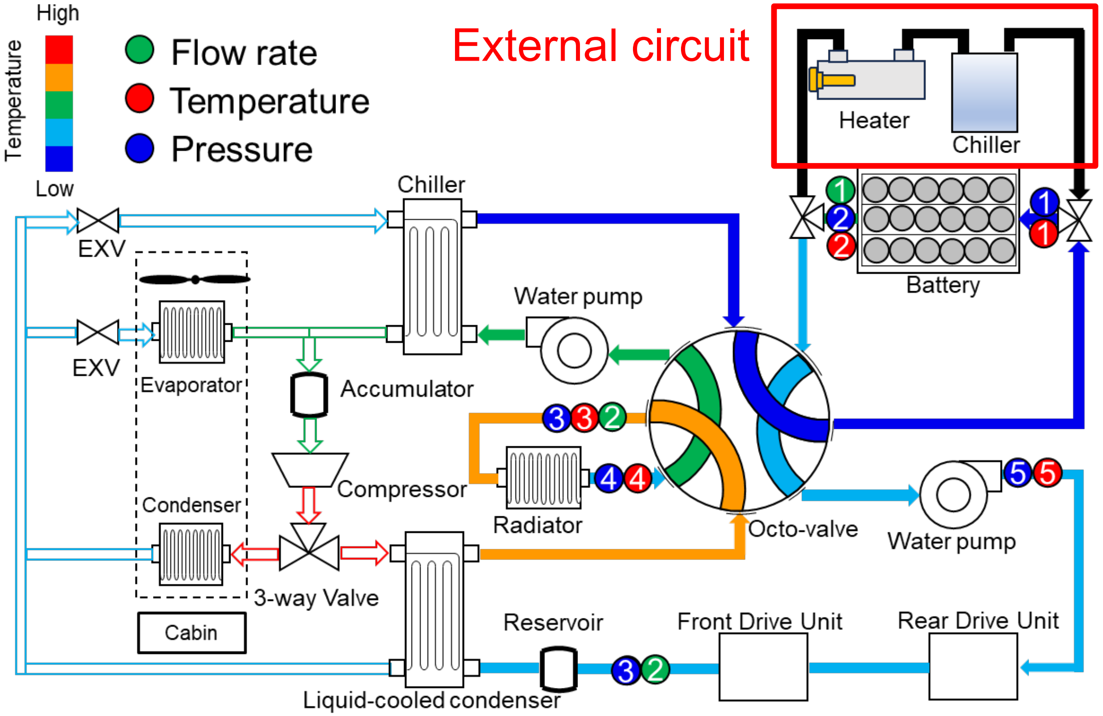
✓ Measured temperature distribution between cells in the direction of cooling water flow

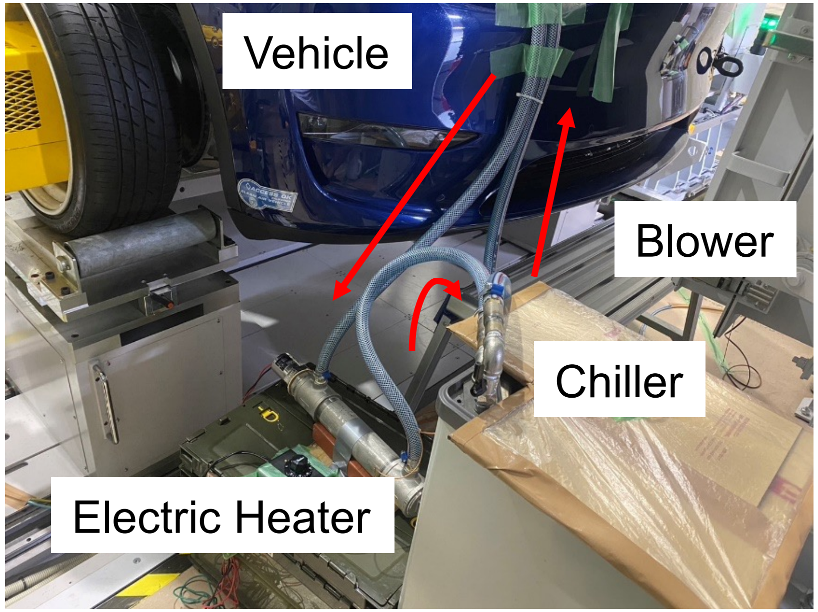




Research method

✓ Additional circuits including a chiller and heater were added to control the initial cell temperature





Temperature condition

Case	Initial battery temperature deg.C	Ambient temperature deg.C
Low	15	18
Middle	25	25
High	40	30

Controlled by additional heat and chiller
Controlled by air conditioner (Min.=18deg.C, Max.= 30deg.C)

Driving cycle test condition

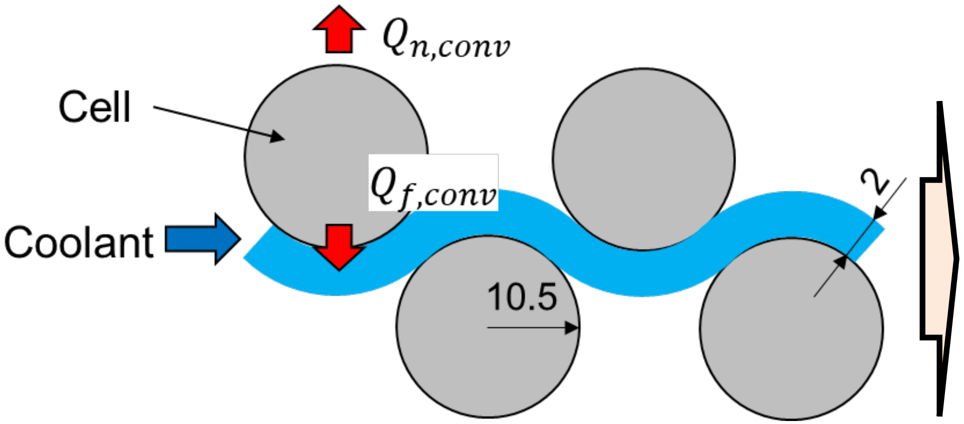
Driving pattern	Temperature case	Initial SOC %
WLTC 2 cycles	Low	20, 50, 80
	Middle	20, 40, 60, 80
	High	20, 50, 80

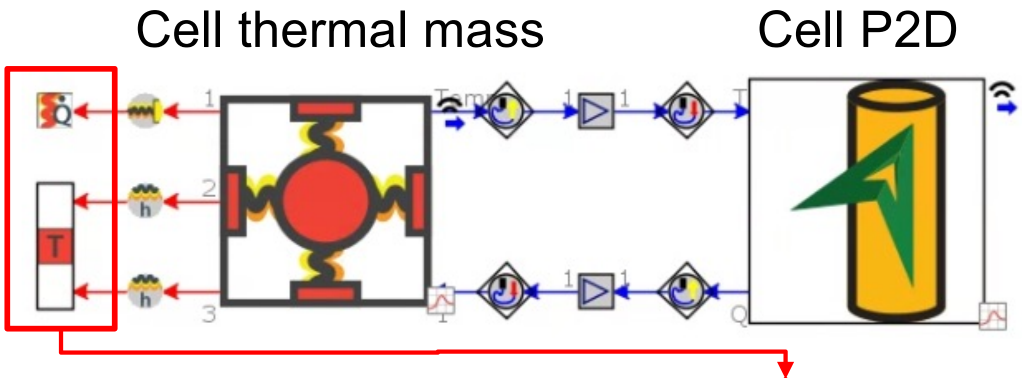
Pressure drop measurement condition

Battery temperature deg.C	Volume flow rate L/min
0, 10, 20, 25, 30, 40	4, 6, 8, 10, 12, 14, 15

Model construction

◆ Overview of model





$$m_{cell}c_{cell}\frac{dT_{cell}}{dt} = Q_{gen} - Q_{f,conv} - Q_{n,conv}$$

$Q_{n,conv}$: Natural convection with ambient
$Q_{f,conv}$: Forced convection with coolant

◆ Model construction method

① Coolant circuit model construction
✓ Pressure drop identification
✓ Heat transfer rate identification

② P2D model construction
✓ Obtain parameters for cathode, anode, electrolyte, etc. from literature

Input heat transfer rate as the boundary conditions

③ P2D model identification
✓ Identify model parameters to reproduce experimental results for cell terminal voltage and temperature

Model construction-Coolant circuit model-

- ✓ Obtain heat transfer rate between cell and coolant for P2D model boundary
- ✓ Identify coolant pressure drop and heat transfer coefficient with coolant circuit model of the entire battery pack

Identify pressure drop coefficient (steady-state)

$$\Delta P = \frac{1}{2} K \rho u^2$$

↓

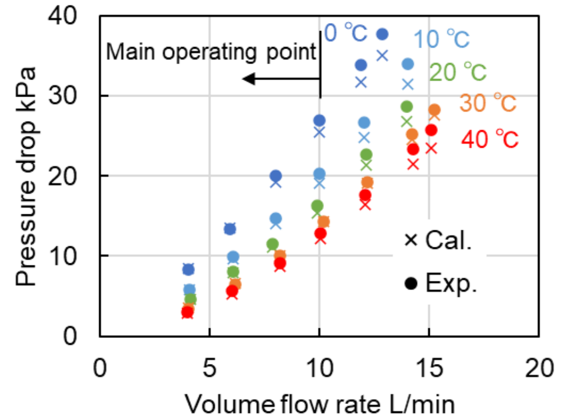
Identify heat transfer coefficient (WLTC)

$$Q_{f,conv} = C_{hm} h_{f,conv} A_{brick} (T_{Brick} - T_{Cool})$$

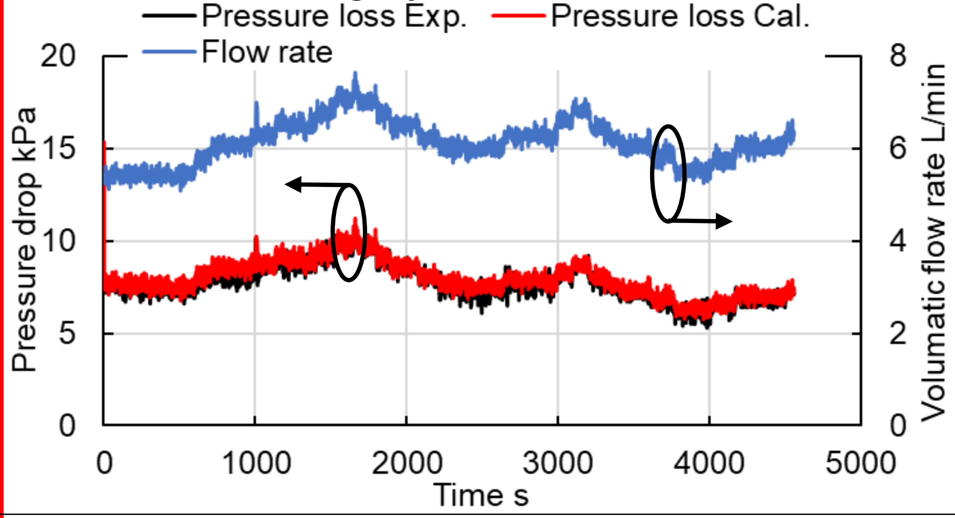
↓

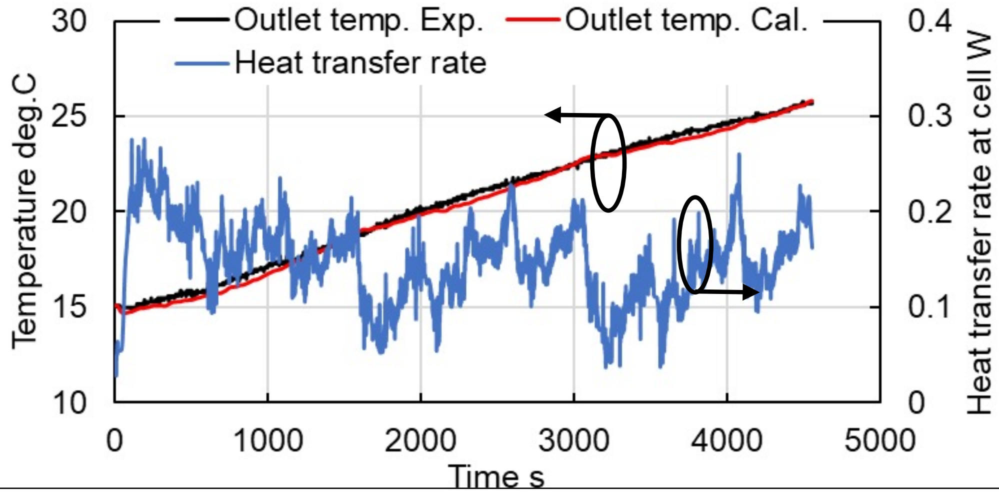
Obtain heat transfer rate

Steady-state test with additional chiller and heater



Driving cycle: WLTC × 2, Initial battery temp: 15deg.C, Initial SOC: 76.6 %

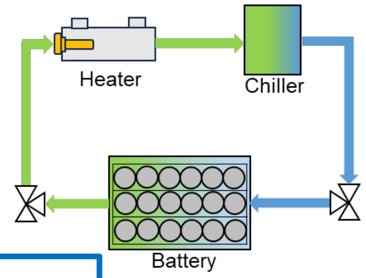




Model construction-Thermodynamics property-

◆ Estimation of heat capacity and natural convection heat transfer coefficient

Using test results of battery cooling test with chiller(40deg.C→20deg.C)



$$C_{bat}\frac{dT_{bat}}{dt} = Q_{f,conv} + Q_{n,conv} \qquad \text{(No heat generation)}$$

$$= \dot{m}_{cool}c_{p,cool}(T_{in} - T_{out}) + h_{n,conv}A_{surf}(T_{bat} - T_{amb})$$

$$T_{bat}^{i} = \frac{1}{C_{bat}}\boxed{\sum(\dot{m}_{cool}c_{p,cool}(T_{in} - T_{out})\Delta t)} + \frac{h_{n,conv}A_{surf}}{C_{bat}}\boxed{\sum((T_{bat} - T_{amb})\Delta t)} + T_{bat}^{0}$$

Calculate $\frac{1}{C_{bat}}$, $\frac{h_{n,conv}A_{surf}}{C_{bat}}$ by multiple regression analysis

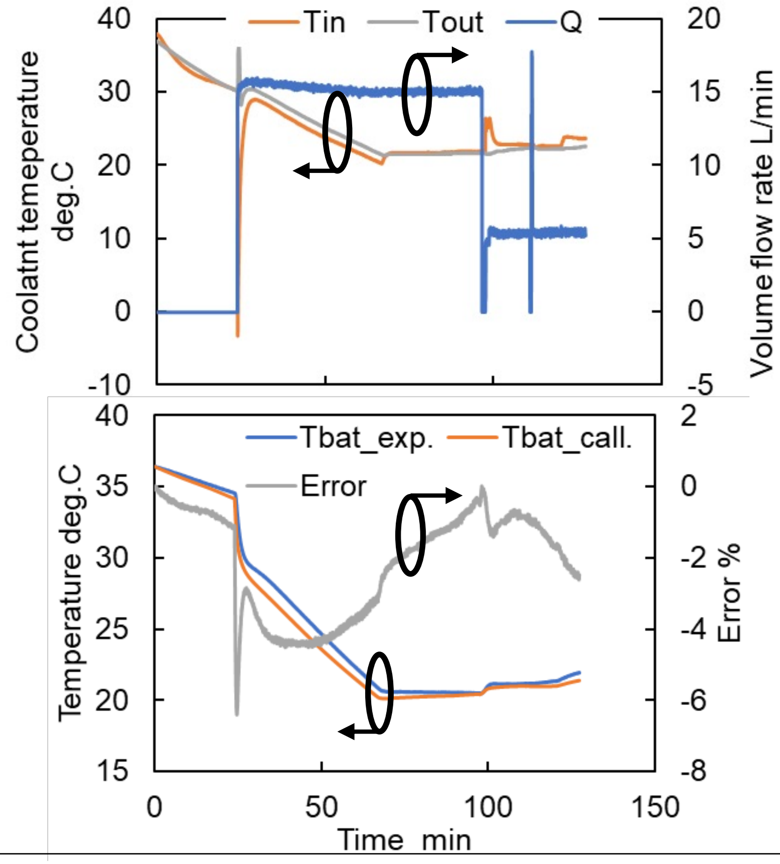
Calculation results of C_{bat} and $h_{n,conv}A_{surf}$

C_{bat} kJ/K	$h_{n,conv}A_{surf}$ W/K	c_{bat} J/(kg･K)	$h_{n,conv}$ W/m²･K
351	33.2	967*	4.80**

* Estimate considering materials other than cells

** Estimate based on battery pack size 95 mm × 1450 mm × 2150 mm

Q_{cool}	W	:Heat dissipation to coolant
$\dot{m}_{cool}$	kg/s	:Coolant mass flowrate
$c_{p,cool}$	J/(kg･K)	:Coolant specific heat capacity
T_{in}	K	:Battery pack inlet coolant temperature
T_{out}	K	:Battery pack outlet coolant temperature
T_{bat}^{i}	K	:Battery pack average temperature at time i
T_{bat}^{0}	K	:Battery pack initial average temperature



Model construction -P2D model-

◆ Chemical reaction at electrode

- ✓ From the literature, it was inferred that the anode was graphite mixed with SiOx
- ✓ The capacity is increased by mixing SiOx

Battery cell specification(5)(6)

Battery model	21700
Diameter x Height mm	20.9 x 70.1
Weight g	68.6
Nominal Capacity Ah	4.60
Energy Wh	16.6
Cathode Material	NCA
Anode Material	SiOx-C

□ Cathode（Positive electrode）

$$LiNi_xCo_yAl_zO_2 \rightleftarrows Li_{1-m}Ni_xCo_yAl_z + mLi^+ + me^-$$

□ Anode（Negative electrode）

$$C_6 + yLi^+ + ye^- \rightleftarrows Li_yC_6$$

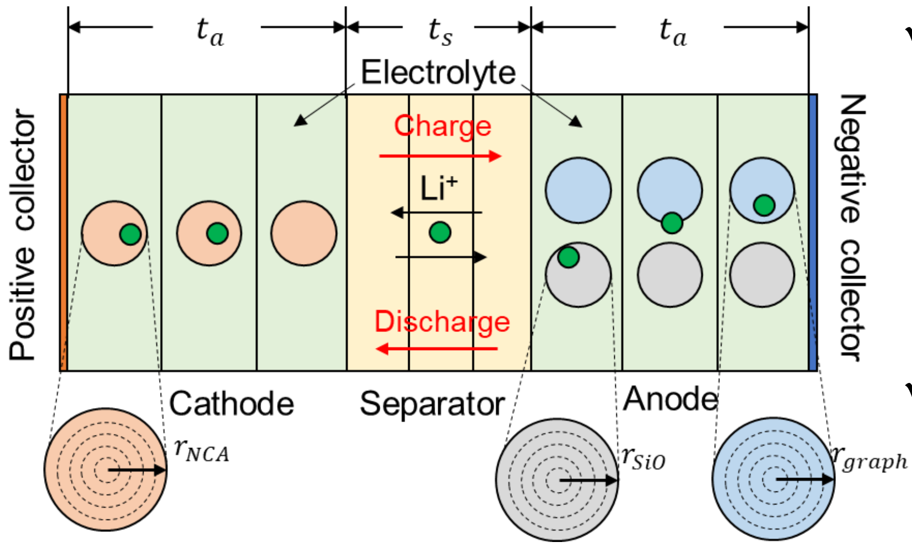
$$SiO + Li \rightarrow \frac{1}{4}Li_4SiO_4 + \frac{3}{4}Si$$ (Irreversible)

$$\frac{1}{4}Li_4SiO_4 + \frac{3}{4}Si + \frac{45}{16}Li^+ + \frac{45}{16}e^+ \rightleftarrows \frac{1}{4}Li_4SiO_4 + \frac{3}{4}Li_{3.75}Si$$
(Reversible)

✓ Mixing SiOx changes the approximate shape of OCV (Open Circuit Voltage) and capacity

Model construction -P2D model-

◆ P2D model(3)(7)



ε		Porosity
c_e	mol/m^3	Molar concentration of lithium ions in the electrolyte
D_e^{eff}	m²/s	Electrolyte effective diffusion coefficient
F	s A/mol	Faraday's constant
t_+^0	-	Transference number
j^{Li}	A	Reaction current of lithium
c_s	mol/m^3	Concentration of lithium in the radial direction of the particles in the active materials
r	m	Radius of the particle
D_s	m²/s	Diffusion coefficient of lithium in the solid phase
φ_e	V	Liquid phase potential
k^{eff}	S/m	Electrolyte's effective ionic conductivity
k_D^{eff}	S/m	Ionic diffusional conductivity

□ Governing equation

✓ Species conservation equation

$$\frac{\partial}{\partial t}[\varepsilon c_e] = \frac{\partial}{\partial x}\left(D_e^{eff}\frac{\partial c_e}{\partial x}\right) + \frac{1-t_+^0}{F}j^{Li} \quad \cdots \text{Liquid}$$

$$\frac{\partial c_s}{\partial t} = \frac{1}{r^2}\frac{\partial}{\partial r}\left(D_s r^2 \frac{\partial c_s}{\partial r}\right) \quad \cdots \text{Solid}$$

✓ Charge conservation equation

$$\frac{\partial}{\partial x}\left(k^{eff}\frac{\partial \varphi_e}{\partial x}\right) + \frac{\partial}{\partial x}\left(k_D^{eff}\frac{\partial \ln c_e}{\partial x}\right) + j^{Li} = 0 \quad \cdots \text{Liquid}$$

$$\frac{\partial}{\partial x}\left(\sigma_e^{eff}\frac{\partial \varphi_s}{\partial x}\right) - j^{Li} = 0 \quad \cdots \text{Solid}$$

✓ Butler-Volmer equation

$$j^{IC} = a_s i_0 \left(e^{\frac{\alpha_a F}{R_u T}\eta} - e^{\frac{\alpha_c F}{R_u T}\eta}\right)$$

$$i_0 = k(C_c)^{a_n}(C_c, max - C_s, surf)^{a_n}\left(C_{c,surf}\right)^{a_p}$$

σ_e^{eff}	S/m	Effective solid-phase conductivity
φ_s		Solid phase potential
a_s	m²/m³	Volume specific reaction surface area
k	A・m$^{2.5}$/mol$^{1.5}$	Reaction rate constant

Model construction -P2D model-

◆ P2D model[(3)(7)]

☐ Heat generation

$$\dot{Q}_{gen} = \dot{Q}_{rx} + \dot{Q}_{rev} + \dot{Q}_{ohm} + \dot{Q}_c$$

✓ Redox reaction heat generation

$$\dot{Q}_{rx} = \frac{1}{L}\int_0^L j^{Li}(\varphi_s - \varphi_e - U)\,dx$$

✓ Ohmic heat generation

$$\dot{Q}_{ohm} = \frac{1}{L}\int_0^L \left[\sigma_e^{eff}\left(\frac{\partial \varphi_s}{\partial x}\right)^2 + k^{eff}\left(\frac{\partial \varphi_e}{\partial x}\right)^2 + k_D^{eff}\left(\frac{\partial ln c_e}{\partial x}\right)\left(\frac{\partial \varphi_e}{\partial x}\right)\right] dx$$

✓ Reversible heat generation

$$\dot{Q}_{rev} = \frac{1}{L}\int_0^L j^{Li}\left(T\frac{\partial U}{\partial T}\right) dx$$

✓ Contact resistance heat generation

$$\dot{Q}_c = \frac{R_c}{A_s V_c} I^2$$

☐ Temperature dependence of parameter

✓ Diffusion coefficient of solid phase

$$D_s = D_{s,ref}\exp\left(\frac{E_D}{R}\left(\frac{1}{T_{ref}} - \frac{1}{T}\right)\right)$$

✓ Reaction rate constant

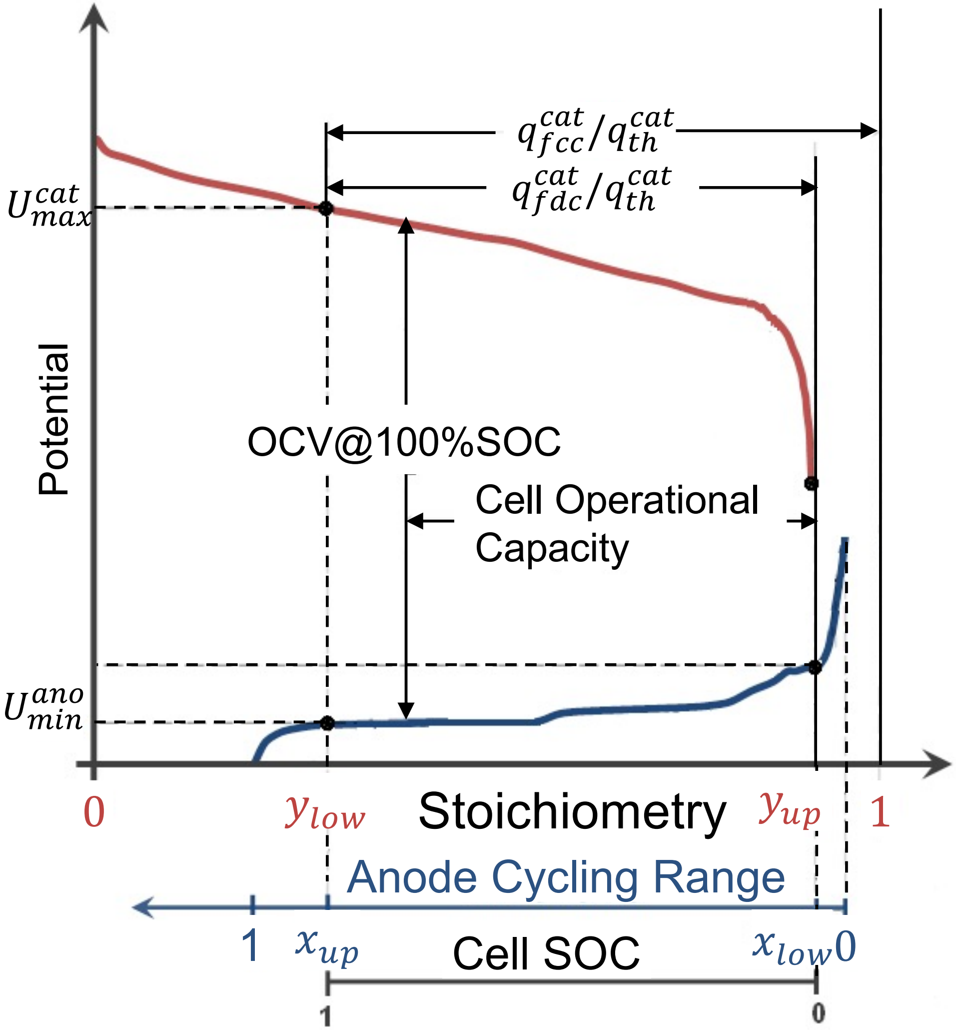
$$k = k_0\exp\left(\frac{E_k}{R}\left(\frac{1}{T_{ref}} - \frac{1}{T}\right)\right)$$

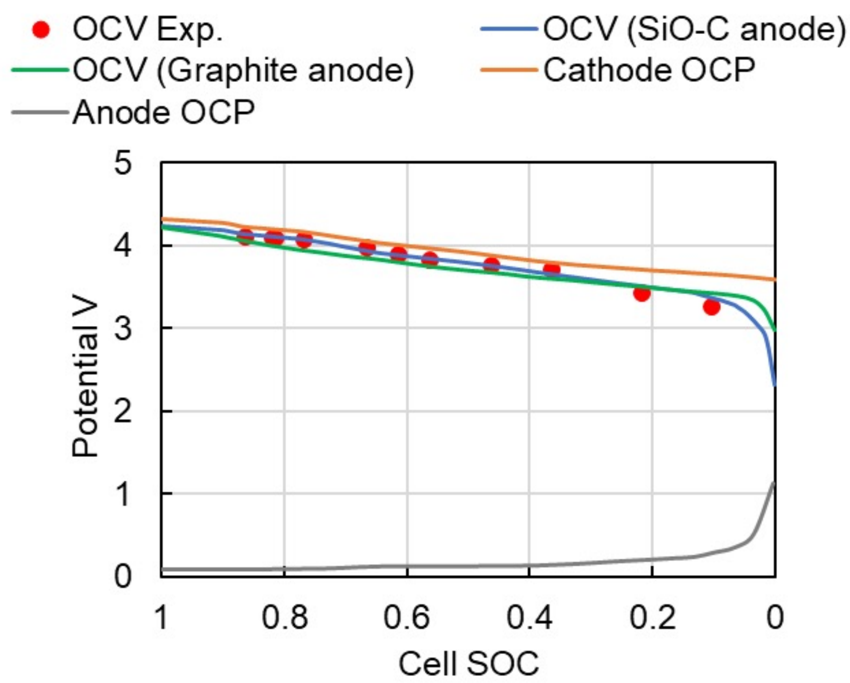
$\dot{Q}_{gen}$	W	: Battery heat generation rate
$\dot{Q}_{rx}$	W	: Redox reaction heat generation rate
$\dot{Q}_{rev}$	W	: Reversible heat generation rate
$\dot{Q}_{ohm}$	W	: Ohmic heat generation rate
$\dot{Q}_c$	W	: Contact resistance heat generation rate
L	m	: Total layer longitudinal thickness
U	V	: Open circuit potential of the solid
A_s	m^2	: Area of contact between solid phase and current collector
V_c	m^3	Cell volume
$D_{s,ref}$	m^2/s	: Diffusion coefficient of at reference temperature
E_{aD_s}	J/mol	: Activation energy for Li diffusion coefficient in solid phase
$i_{0,ref}$	m^2/s	: Exchange current density at reference temperature
E_{ai_0}	J/mol	: Activation energy for exchange current density
T_{ref}	K	: Reference temperature

Model calculation

◆ Cell balancing

- ✓ OCV is defined by cathode and anode OCP(open circuit potential) balance
- ✓ OCP vs Stoichiometry curve is obtained from the literature
- ✓ Parameters that define cell balance were identified before transient calculation

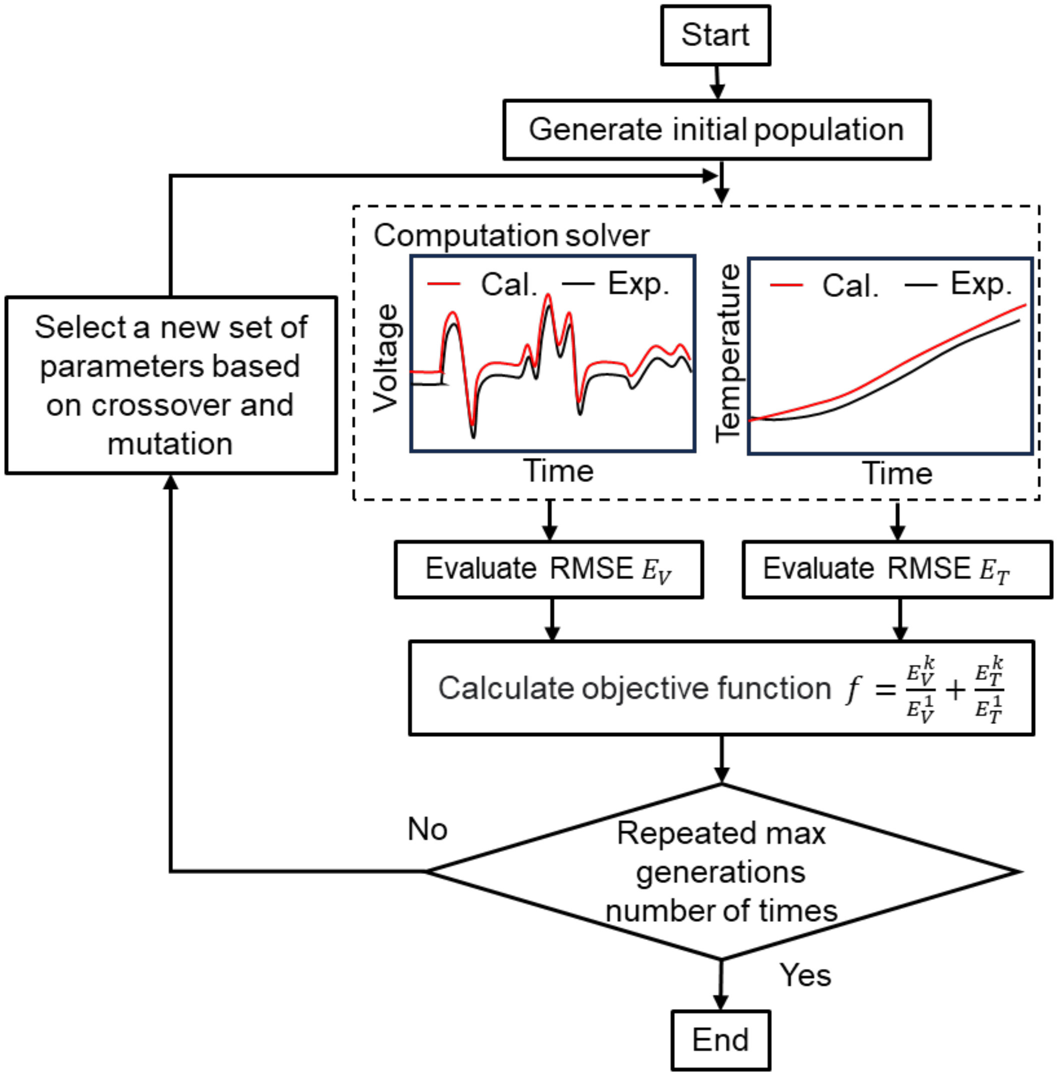




✓ OCV is closer to experimental values than for graphite-only anode

Model construction -P2D model-

◆ Parameter identification flow



□ Calculation conditions

Experimental data used for parameter identification

Case	Experimental data	Temperature Case	Initial SOC %
1	WLTC 2 cycles	Low	76.6
2		Middle	36.4
3		Middle	61.3
4		High	21.4

□ Objective functions

$$E_V = \sqrt{\frac{\int_{t_{start}}^{t_{end}} \left(V(t) - V(t)_{target}\right)^2 dt}{(t_{end} - t_{start})}}$$

$$E_T = \sqrt{\frac{\int_{t_{start}}^{t_{end}} \left(T(t) - T(t)_{target}\right)^2 dt}{(t_{end} - t_{start})}}$$

$$f = \frac{E_V^k}{E_V^1} + \frac{E_T^k}{E_T^1}$$

Model construction -P2D model-

◆ Identified parameters

Each parameter was set within the range of the minimum and maximum values obtained in the literature.

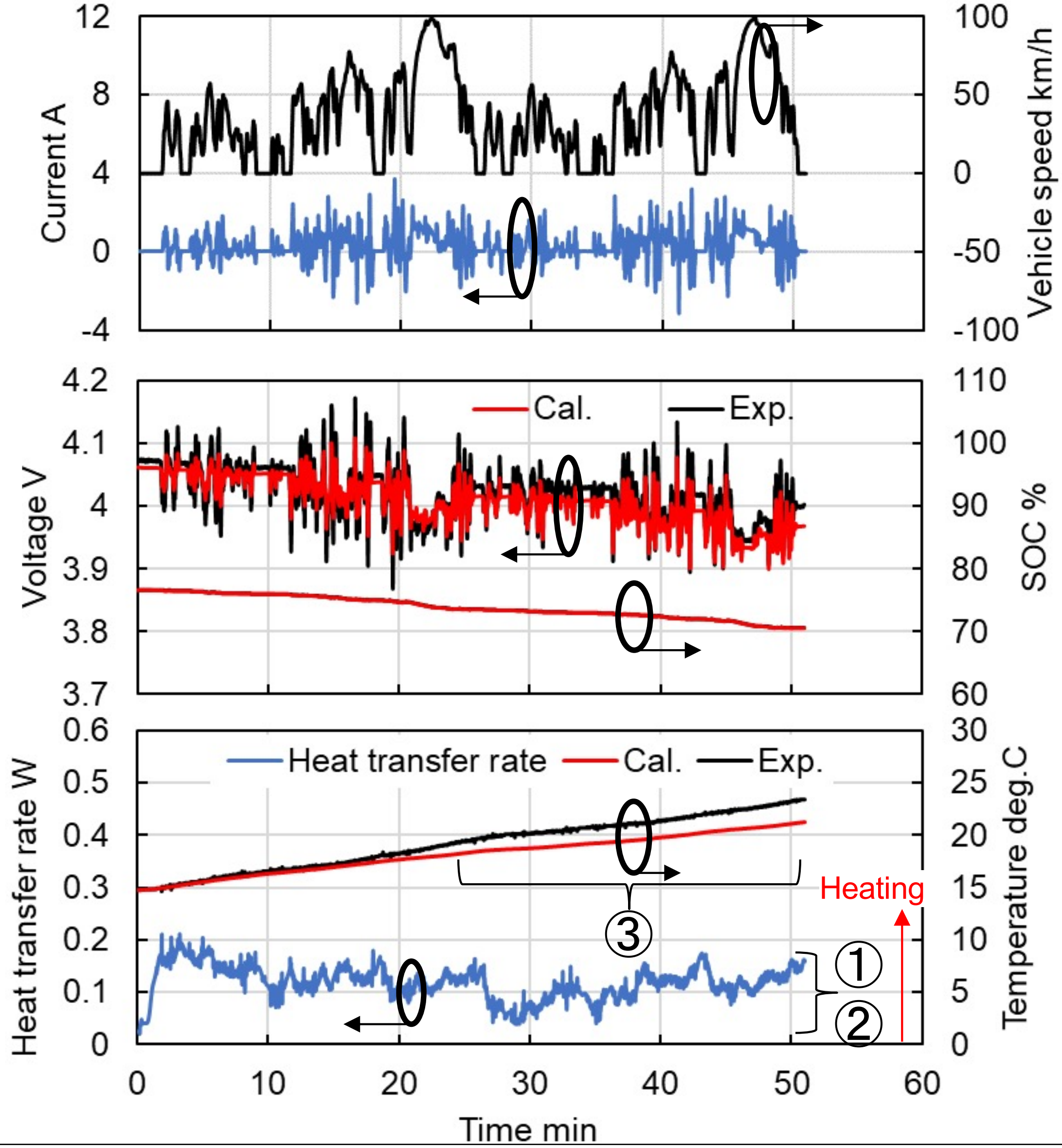
Identified parameters

Parameter	Symbol	Unit	Cathode	Separator	Anode	
			NCA		C_6	SiO
Thickness	$t_{cat}, t_{sep}, t_{ano}$	μm	94	13	96	
Particle size	r_{NCA}, r_C, r_{SiO}	-	3.6	-	18	9.7
Reaction rate constant	$k_{0,NCA}, k_{0,C}, k_{0,SiO}$	$m^{2.5}/(mol^{0.5}s)$	9.6×10^{-8}	-	1.3×10^{-9}	1.4×10^{-12}
Diffusion coefficient of solid	$D_{s,NCA}, D_{s,C}, D_{s,SiO}$	m^2/s	2.1×10^{-14}	-	1.1×10^{-13}	8.9×10^{-12}
Activation energy of diffusion coefficient	$E_D^{NCA}, E_D^C, E_D^{SiO}$	kJ/mol	1.8	-	47	17
Activation energy of reaction rate constant	$E_k^{NCA}, E_k^C, E_k^{SiO}$	kJ/mol	20	-	8.6	13
Contact Resistance	R_{cat}, R_{ano}	$\Omega\cdot m^2$	8.6×10^{-4}	-	8.6×10^{-4}	
Porosity	$\varepsilon_{cat}, \varepsilon_{sep}, \varepsilon_{ano}$	-	0.20	-	0.21	
First charge capacity	$q_{fcc}^{NCA}, q_{fcc}^C, q_{fcc}^{SiO}$	mAh/g	225	-	372	2592
First discharge capacity	$q_{fdc}^{NCA}, q_{fdc}^C, q_{fdc}^{SiO}$	mAh/g	206	-	329	1842
SiO mass fraction	m_{SiO}	-	-	-	-	0.040
Maximum potential	U_{max}^{cat}	V	4.2	-	-	-
OCV at SOC 100 %	U_{100}^{cell}	V	4.2			
OCV at SOC 0 %	U_0^{cell}	V	2.3			

Model calculation

◆ Parameter identification results(Case1: Low temperature)

Initial cell temperature: 14.8deg.C, Initial SOC: 76.6%



□ Experimental results

① Water pump never stopped after turning on the vehicle

② Battery cells were heated throughout the test.

□ P2D model results

③ Temperature calculation accuracy deteriorated after 20 minutes.

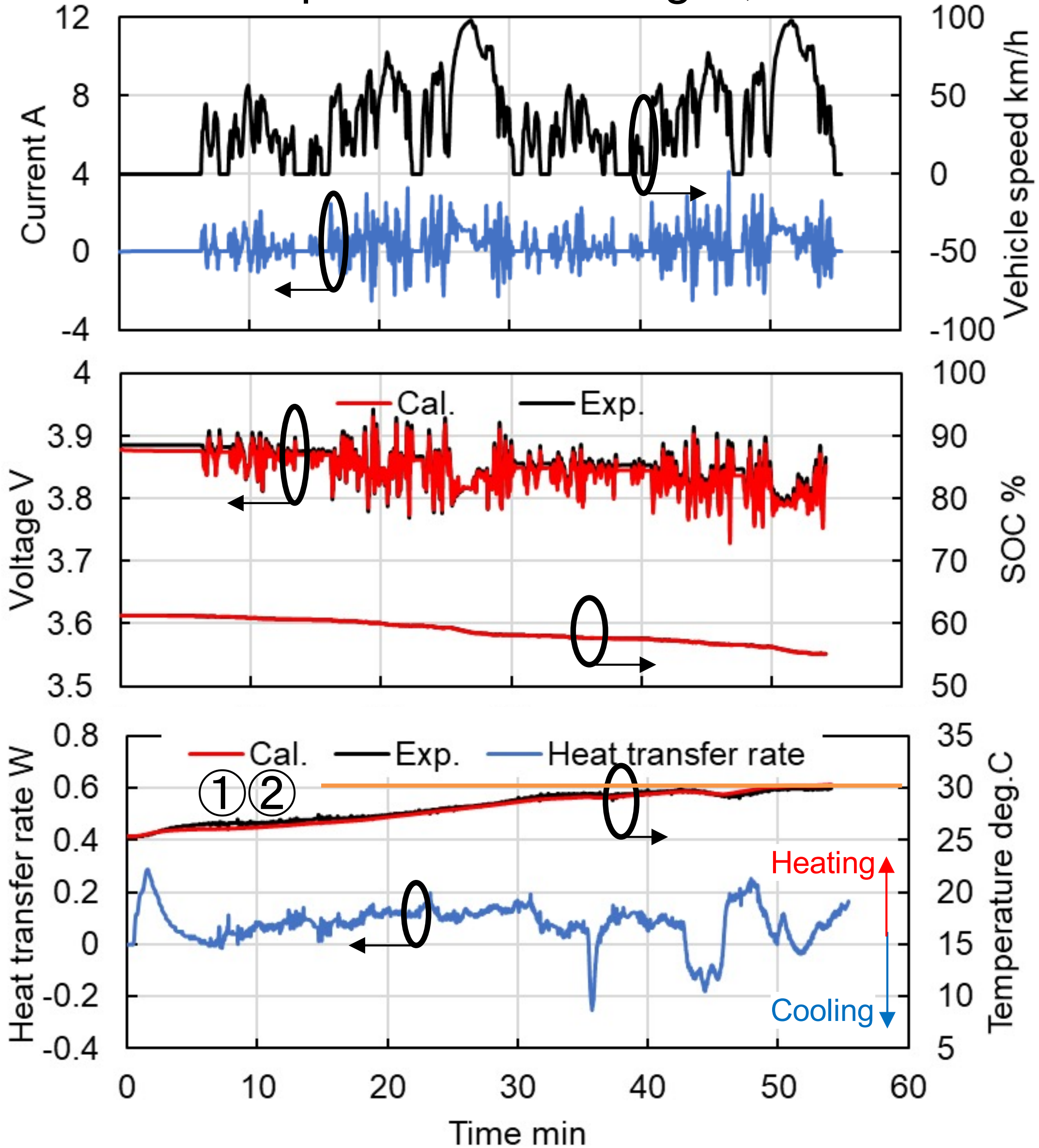
Model calculation results

	Voltage	Temperature
RMSE	0.019 V	1.2deg.C
Max. error rate	1.5%	9.8%
Max error @ battery pack Cell voltage × 96	6.1 V	-

Model calculation

◆ Parameter identification results(Case3: Middle temperature)

Initial cell temperature: 25.3deg.C, Initial SOC: 61.3%



□ Experimental results

① The battery was heated with coolant until it reached around 30deg.C (around 35 min)

② After heating cell temperature was maintained around30 deg.C

□ P2D model results

③ Voltage and temperature were calculated with high accuracy.

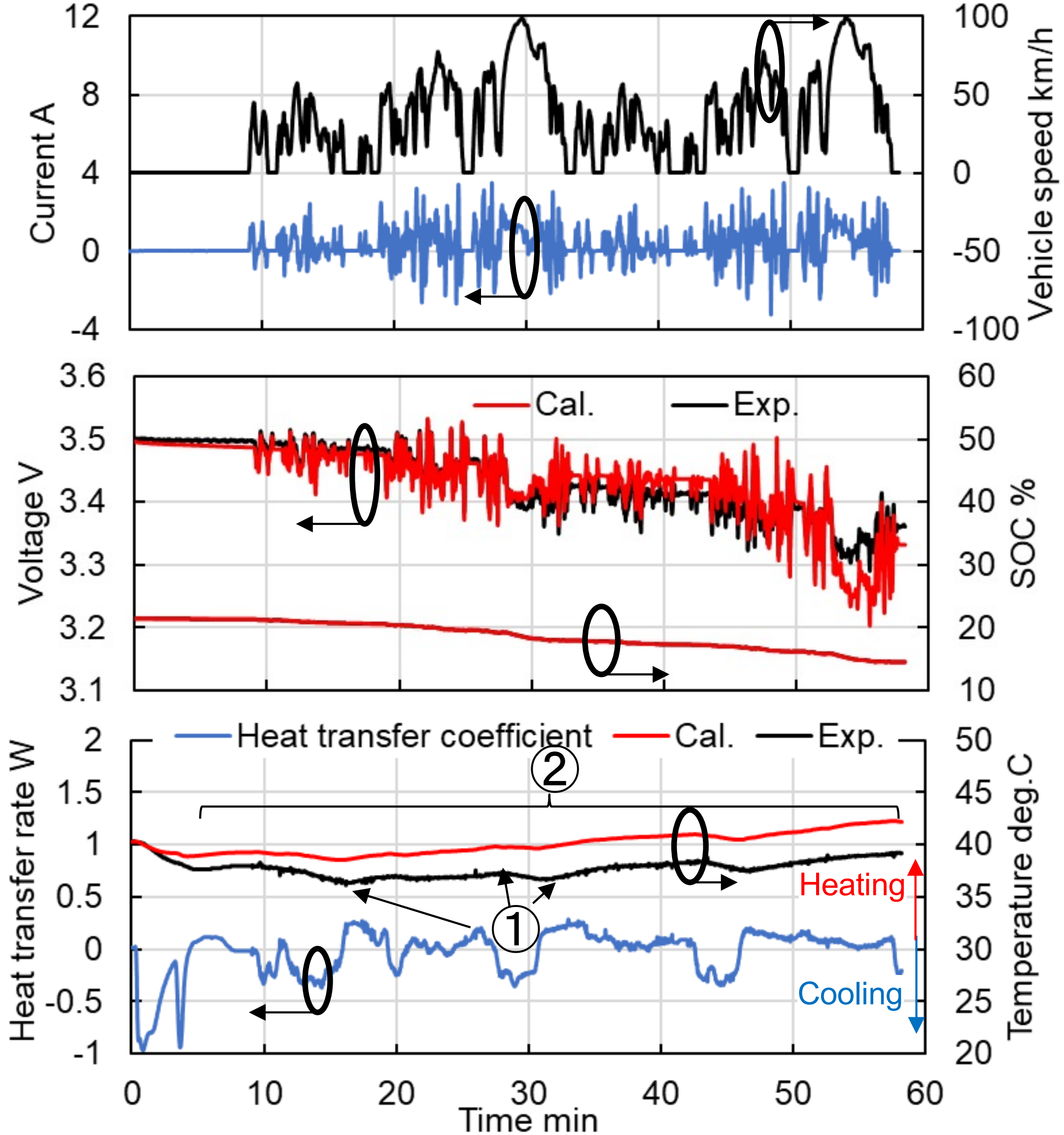
Model calculation results

	Voltage	Temperature
RMSE	0.0072V	0.28deg.C
Max. error rate	0.16%	2.1%
Max error @ battery pack Cell voltage×96	1.6 V	-

Model calculation

◆ Parameter identification results(Case4: High temperature)

Initial cell temperature: 40.3deg.C, Initial SOC: 21.4%



☐ Experimental results

① Battery cell temperature maintained at 37~39deg.C

☐ P2D model results

② Low accuracy of temperature calculations throughout the calculation

→Heat generation is largely calculated at low SOC

Model calculation results

	Voltage	Temperature
RMSE	0.022V	2.5deg.C
Max. error rate	2.7%	9.1%
Max error @ battery pack Cell voltage × 96	8.4V	-

Model calculation analysis

◆ Problem of heat generation at low SOC

$$\dot{Q}_{gen} = \underbrace{\dot{Q}_{rx} + +\dot{Q}_{ohm} + \dot{Q}_c}_{} + \dot{Q}_{rev}$$

$$\dot{Q}_{gen} = \boxed{I(V_{OCV} - V_t)} + IT\frac{dV_{OCV}}{dT}$$

Total heat generation due to overvoltage matches if terminal voltage calculations match

⇩

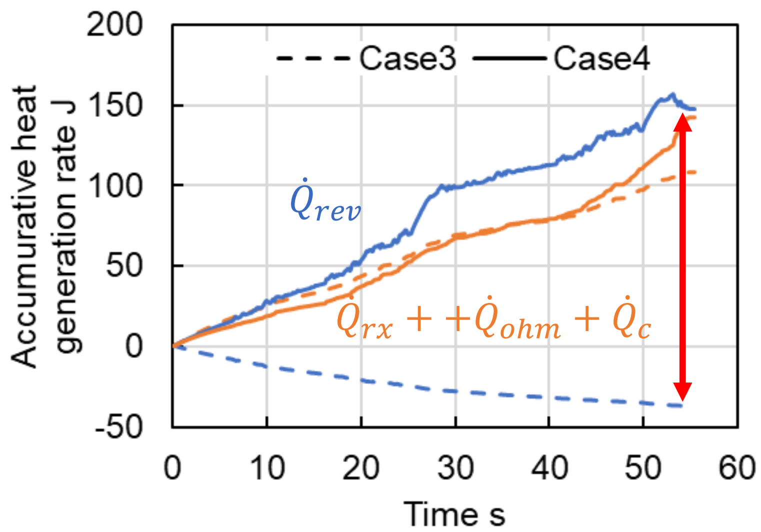
Reversible heat generation $\dot{Q}_{rev}$ is the cause of the temperature error.

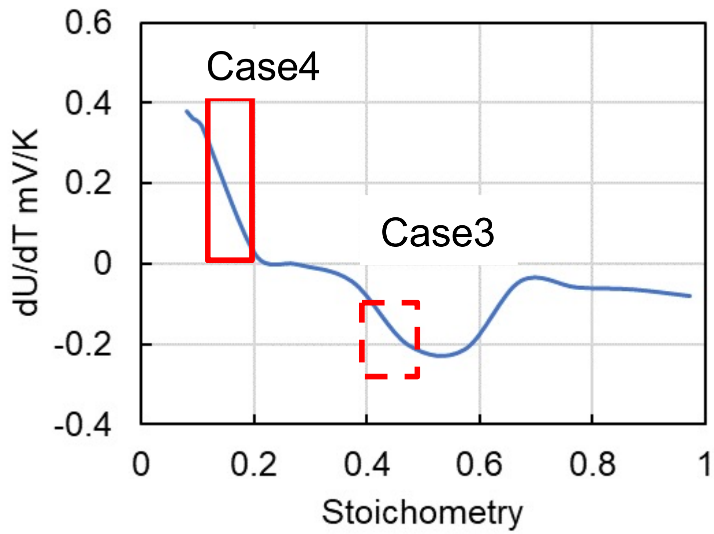
⇩

One of the following improvements is needed

✓ SiO-C anode dU/dT map

✓ Cell balancing of anode and cathode





(8) Kieran O'Regan et al., Elsevier, 2022

Conclusion

In this study, we tried to construct a battery P2D model based on vehicle driving tests. As a result, the following findings were obtained.

- By adding a heater and chiller circuit outside the vehicle, it was possible to measure the pressure drop and identify the model under steady-state conditions. It was also possible to estimate the heat capacity and natural convection heat transfer coefficient.

- P2D model identification of battery cells using vehicle driving test results is possible by modeling heat dissipation from battery cells to cooling water and to the atmosphere.

- The model can be calculated with an error of up to 2.7 % for voltage and 9.8 % for temperature.

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