# Supporting Information

## Decouple Charge Transfer Reactions in the Li-ion Battery

### S1. Deduction of Models A–F for LIBs with the complex phasor method

This section gives how to deduce Models A–F based on Model DFN.

#### S1.1. Deduction of Model A and Model D

Eqs. (1-1)–(45) are still applicable to Model A, and the differences will be described as follows.

In the case of *D*e,eff3→∞, substituting Eq. (45) into BCs of Eqs. (38-1) and (38-2) yields

Substituting Eq. (S1-1) into Eq. (37-2) and integrating the newly obtained equation, the integration result can be combined with Eq. (41) to yield

Till now, the solution expressions of (i = 1, 2, 3) are known. However, among them both and are still unknown. The continuity BCs of Eqs. (39-1) and (39-2) are used to evaluate and .

Substituting , , , and according to Eqs. (42-1), (42-5) and (S1-1) into Eqs. (39-1) and (39-2) yields

where the coefficients ΛI1, ΛII1, ΛI2, and ΛII2 are given as

Till now, , , , , , and are all solved. With all these variables solved, , , , , , and can all be obtained in reverse with respect to Eqs. (33-1)–(33-6) and (36-1)–(36-5). Herein, the dimensionless variables and are used and the transfer functions of , , and can be shown as follows

Next, the solid-phase potential in the electrode will be solved. Transforming Eq. (4-1) into the complex phasor field and combining the result with Eq. (23-3) to yield

The BC of Eq. (4-2) combined with Eq. (13-2) can be rewritten in the complex phasor form

Based on Eqs. (33-2)–(33-4), Eqs. (S5-1) and (S5-2) can be rewritten in the dimensionless form

combined with the continuity BC of Eq. (40-1) to yield

Among them, Eqs. (S5-1)–(S6-3) are consistent with Eqs. (52-1)–(53-3) in the text.

Upon integrating Eq. (S6-1) twice and combining Eq. (42-2), the transfer function of is evaluated as follows

Using the gradient BC of Eq. (S6-2), *H*I1 is evaluated to be

Using Eq. (S6-3), *H*II1 is evaluated to be

where *Γ*I1 and *Γ*II1 are given as

The results are similar to Eqs. (S7-1)–(S7-3) can be obtained for the positive electrode.

The analytical expression (66-1)–(66-4) of Model A can be finally obtained by combining Eqs. (56-1)–(56-4) with Eqs. (S4-1)–(S4-3), (S7-1), and (S8-1).

Here, the impedance expression of Model D can be obtained by setting *z*d = 0 in Eqs. (23-1)–(23-3) and modifying the coefficients of Eqs. (66-1)–(66-4).

#### S1.2. Deduction of Model B and Model E

Eqs. (1-1)–(41) are still applicable to Model B, and the differences will be described as follows.

Eqs. (35-1) and (35-3) can be rewritten in a more concise form

In the case of *D*e,eff→∞, solving Eqs. (34-1) and (34-3) with Eqs. (S9-1) and (S9-2) can yield

In the case of *D*e,eff3→∞, solving Eq. (37-1) with Eqs. (38-1) and (38-2) can yield

In the case of *D*e,eff→∞, combining Eqs. (S10-1) and (S10-2) with Eqs. (34-2) and (34-4) can yield

The general solution of Eqs. (S11-1) and (S11-2) can be obtained directly

Eqs. (35-2) and (35-4) combined with Eqs. (S10-1) and (S10-2) yields

By substituting Eqs. (S12-1) and (S12-2) into BCs of Eqs. (S13-1) and (S13-2), the coefficients *B*III and *B*IV can be obtained.

For

For

Substituting Eq. (S10-3) into Eq. (37-2) and integrating the newly obtained equation, the integration result can be combined with Eq. (41) to yield

Eq. (S15) is similar to Eq. (S1-2).

Till now, , , , , , and are all solved. With all these variables solved, , , , , , and can all be obtained in reverse with respect to Eqs. (33-1)–(33-3) and (36-1)–(36-3). Herein, the dimensionless variable is used and the transfer functions of , , and can be shown as follows

Next, the solid-phase potential in the electrode will be solved. Transforming Eq. (4-1) into the complex phasor field and combining the result with Eq. (23-3) to yield

The BC of Eq. (4-2) combined with Eq. (13-2) can be rewritten in the complex phasor form

Among them, Eqs. (S17-1) and (S17-2) are consistent with Eqs. (52-1) and (52-2) in the text.

Based on Eqs. (33-2)–(33-3), Eqs. (S17-1) and (S17-2) can be rewritten in the dimensionless form

combined with the continuity BC of Eq. (40-1) to yield

Among them, Eqs. (S18-1)–(S18-3) are similar to Eqs. (53-1)–(53-3) in the text.

Upon integrating Eq. (S18-1) twice and combining Eq. (S12-1), the transfer function of is evaluated as follows

Using the gradient BC of Eq. (S18-2), *H*I1 is evaluated to be

Using Eq. (S18-3), *H*II1 is evaluated to be

where *Γ*II1 is given as

The results are similar to Eqs. (S19-1)–(S19-3) can be obtained for the positive electrode

The analytical expression (74-1)–(74-4) of Model B can be finally obtained by combining Eqs. (56-1)–(56-4) with Eqs. (S16-1)–(S16-3), (S19-1), and (S20-1).

Here, the impedance expression of Model E can be obtained by setting *z*d = 0 in Eqs. (23-1)–(23-3) and modifying the coefficients of Eqs. (74-1)–(74-4).

#### S1.3. Deduction of Model C and Model F

Eqs. (1-1)–(32) are still applicable to Model C, and the differences will be described as follows.

In the case of *D*e,eff→∞, Eqs. (25-1) and (25-3) combined with Eqs. (26-1)–(26-4) can yield

In the case of *D*e,eff3→∞, Eq. (28-1) combined with Eqs. (29-1) and (29-2) can yield

Substituting Eq. (S21-3) into Eq. (28-2) and integrating the newly obtained equation, the integration result can be combined with Eq. (32) to yield

In the case of *D*e,eff→∞ and *σ*eff→∞, Eqs. (25-2) and (25-4) combined with Eqs. (S21-1) and (S21-2) can yield

In the case of *D*e,eff→∞ and *σ*eff→∞, Eqs. (27-1)–(27-4) combined with Eqs. (S21-1) and (S21-2) can yield

To rewrite the above GEs with IBCs in a more concise form and to follow the previous method, the dimensionless variables are defined as

Based on Eqs. (S25-1)–(S25-3), Eqs. (S23-1)–(S24-4) can be rewritten in the dimensionless form

The general solutions of Eqs. (S26-1) and (S26-2) can be resolved directly

By substituting Eqs. (S28-1) and (S28-2) into BCs of Eqs. (S27-1)–(S27-4), the coefficients *B*III and *B*IV can be obtained.

For

For

Till now, and are all solved. With all these variables solved, and can be obtained in reverse with respect to Eqs. (S25-1)–(S25-3). Herein, the dimensionless variables is used and the transfer functions of and can be shown as follows

Next, the solid-phase potential in the electrode will be solved. Transforming Eq. (4-1) into the complex phasor field can yield

The BC of Eq. (4-2) combined with Eq. (13-2) can be rewritten in the complex phasor form

Among them, Eqs. (S31-1) and (S31-2) are consistent with Eqs. (52-1) and (52-2) in the text.

In the case of *σ*eff1→∞, Eqs. (S31-1) and (S31-2) can be rewritten as

combined with the continuity BC of Eq. (31-1) to yield

Upon integrating Eq. (S32-1) twice, the transfer function of is evaluated as

By substituting Eq. (S33-1) into BCs of Eqs. (S32-2) and (S32-3), the coefficients *H*I1 and *H*II1 can be obtained

where *Γ*II1 is given as

The results are similar to Eqs. (S33-1)–(S33-3) can be obtained for the positive electrode

The analytical expression (78-1)–(78-4) of Model C can be finally obtained by combining Eqs. (56-1)–(56-4) with Eqs. (S22), (S30-1), (S30-2), (S33-1) and (S34-1).

Here, the impedance expression of Model D can be obtained by setting *z*d = 0 in Eqs. (23-1)–(23-3) and modifying the coefficients of Eqs. (78-1)–(78-4).

To sum up, although simplified impedance models such as Models A–F still have: (i) the electrolyte phase concentration flux continuity BCs of Eqs. (1-3) and (6-2); (ii) the electrolyte phase concentration continuity BCs of Eqs. (11-2) and (11-3); (iii) the electrolyte phase current density continuity BCs of Eqs. (2-3) and (7-2); and (iv) the electrolyte phase potential continuity BCs of Eqs. (12-2) and (12-3) at the electrode/separator interface, the electrode impedance can be calculated independently.

### S2. Unification the DFN-like impedance models

This section gives how to degenerate the analytical expression Eqs. (57-1)–(57-4) of Model DFN into Eqs. (66-1)–(66-4) of Model A or Model D, then from Eqs. (66-1)–(66-4) of Model A or Model D to Eqs. (74-1)–(74-4) of Model B or Model E, and finally from Eqs. (74-1)–(74-4) of Model B or Model E to Eqs. (78-1)–(78-4) of Model C or Model F.

#### S2.1. Degeneration from Model DFN to Model A & D

In case of *D*e,eff3→∞, the coefficients ΛIII3, ΘIII3, *Γ*III3, *B*I3 and *B*II3 in Eqs. (60-5), (63), (65-1)–(65-3) tend to zero, resulting , , and *H*II1 in Eqs. (59-5), (62-5), and (64-2) are rewritten as Eqs. (68-5), (71-5), and (73-2), respectively. Therefore, the expressions of Eqs. (57-1)–(57-2) for electrode impedance *Z*1 & *Z*2 and the expressions of Eq. (57-4) for cell impedance *Z*4 remain unchanged, but their internal coefficients change. In addition, the separator impedance *Z*3 of Eq. (57-3) is rewritten as Eq. (66-3).

The degradation from Model A to Model D can be found in section S1.1.

#### S2.2. Degeneration from Model A to Model B & E

In the case of *D*e,eff→∞, the electrolyte-related diffusion resistance is negligible compared with the electrode-related ohmic resistance, that is, ΘI << ΘII. As a result, the eigenvalues *λ*I and *λ*II in Eqs. (68-3) & (68-4) and (71-3) & (71-4) are reduced to

In addition, the concentration flux in Eqs. (68-5) and (71-5) are reduced to

Substituting Eqs. (S35-1), (S35-2) and (S36-1) into Eqs. (67-5) and (67-6) yield *B*III1 and *B*IV1

Substituting Eqs. (S35-3), (S35-4) and (S36-2) into Eqs. (70-5) and (70-6) yields *B*III2 and *B*IV2

Plugging in the values of the coefficients in Eqs. (S35-1), (S35-2), (S37-1), and (S37-2) into the impedance expression (66-1) yields

Plugging in the values of the coefficients in Eqs. (S35-3), (S35-4), (S37-3), and (S37-4) into the impedance expression (66-2) yields

Eqs. (S38-1) and (S38-2) can be expressed in a more concise form as Eqs. (74-1) and (74-2). In addition, the simplified steps from Eq. (66-4) to Eq. (74-4) for the full cell impedance are similar and will not be repeated here.

The degradation from Model B to Model E can be found in section S1.2.

#### S2.3. Degeneration from Model B to Model C & F

In case of *σ*eff→∞, *κ*eff << *σ*eff. As a result, the dimensionless parameter ΘII in Eqs. (75-1) and (75-2) are reduced to

It can be obtained from the electrode impedance Eqs. (74-1) and (74-2) of Model B in case of *σ*eff→∞

Eqs. (S40-1) and (S40-2) can be expressed in a more concise form as Eqs. (78-1) and (78-2). In addition, the simplified steps from Eq. (74-4) to Eq. (78-4) for the full cell are similar and will not be repeated here.

The degradation from Model C to Model F can be found in section S1.3.

### S3. List of abbreviations and symbols

#### S3.1. Abbreviations

1D: one-dimensional

3D: three-dimensional

DFN: Doyle-Fuller-Newman

ECM: equivalent circuit model

EIS: electrochemical impedance spectroscopy

GE: governing equation

IBC: initial boundary condition

LIB: Li-ion battery

ocp: open circuit potential

P2D: pseudo-2-dimensional

sei: solid electrolyte interlay at the particle scale

SEI: solid electrolyte interlay at the electrode scale

SOC: state of charge

TLM: transmission line model

#### S3.2. Latin symbols

*A*: surface area of the electrode, m2

*a*s1: specific surface area of the negative electrode is defined as , m2 m–3

*a*s2: specific surface area of the positive electrode is defined as , m2 m–3

brug1: Bruggeman coefficient in the negative electrode

brug2: Bruggeman coefficient in the positive electrode

brug3: Bruggeman coefficient in the separator

*C*dl1: double layer capacitance of the negative electrode at the particle scale, F m–2

*C*dl2: double layer capacitance of the positive electrode at the particle scale, F m–2

*C*DL1: double layer capacitance of the negative electrode at the electrode scale, F m–2

*C*DL2: double layer capacitance of the positive electrode at the electrode scale, F m–2

*c*e1(*x*,*t*): electrolyte Li+ concentration in the negative electrode, mol m–3

*c*e2(*x*,*t*): electrolyte Li+ concentration in the positive electrode, mol m–3

*c*e3(*x*,*t*): electrolyte Li+ concentration in the separator, mol m–3

: debiased version of *c*e(*x*,*t*) is defined as , mol m–3

: complex phasor for , mol m–3

: dimensionless electrolyte Li+ concentration in the negative electrode is defined as

: dimensionless electrolyte Li+ concentration in the positive electrode is defined as

: dimensionless electrolyte Li+ concentration in the separator is defined as

*c*e,0: Li+ initial concentration in the electrolyte phase*,* mol m–3

*c*s1(*x*,*r*,*t*):solid phase Li concentration in the negative electrode,mol m–3

*c*s2(*x*,*r*,*t*):solid phase Li concentration in the positive electrode,mol m–3

: debiased version of *c*s(*x*,*r*,*t*) is defined as , mol m–3

: complex phasor for , mol m–3

*c*s1,0: Li initial concentration in the negative electrode particle is defined as *,* mol m–3

*c*s2,0: Li initial concentration in the positive electrode particle is defined as *,* mol m–3

*C*sei1: sei film capacitance in the negative electrode at the particle scale is defined as , F m–2

*C*sei2: sei film capacitance in the positive electrode at the particle scale, F m–2

*C*SEI1: SEI film capacitance in the negative electrode at the electrode scale, F m–2

*C*SEI2: SEI film capacitance in the positive electrode at the electrode scale, F m–2

*c*s1,max: maximum Li concentration in the negative electrode particle*,* mol m–3

*c*s2,max: maximum Li concentration in the positive electrode particle*,* mol m–3

*c*ss(*x*,*t*): Li concentration in the particle surface is defined as , mol m–3

: debiased version of *c*ss(*x*,*t*) is defined as , mol m–3

: complex phasor for , mol m–3

*D*e: Li+ diffusion coefficient in the electrolyte phase, m2 s–1

*D*e,eff1: effective electrolyte diffusion coefficient in the negative electrode is defined as , m2 s–1

*D*e,eff2: effective electrolyte diffusion coefficient in the positive electrode is defined as , m2 s–1

*D*e,eff3: effective electrolyte diffusion coefficient in the separator is defined as , m2 s–1

*D*s1: Li solid phase diffusion coefficient in the negative electrode, m2 s–1

*D*s2: Li solid phase diffusion coefficient in the positive electrode, m2 s–1

*F*: Faraday’s constant, 96487 C mol–1

*E*De: activation energy for electrolyte diffusion, J mol–1

*E*Ds1: activation energy for solid phase diffusion in the negative electrode, J mol–1

*E*Ds2: activation energy for solid phase diffusion in the positive electrode, J mol–1

*E*k1: activation energy for reaction constant in the negative electrode, J mol–1

*E*k2: activation energy for reaction constant in the positive electrode, J mol–1

*E*κ: activation energy for electrolyte conductivity, J mol–1

*E*ρsei1: activation energy for sei resistivity in the negative electrode, J mol–1

*f*: frequency, Hz

*f*±: mean molar activity coefficient

*i*(*t*): applied (dis) charge current density, where *i* > 0 indicates discharging, A m–2

: phasor of complex sinusoidal current density signal *i*(*t*), A m–2

*i*1,0(*x*,0): exchange current density for the insertion process in the negative electrode is defined as , A m–2

*i*app(*t*): applied (dis) charge current, A

: electrochemical double-layer current density in the negative electrode is defined as , A m–2

*i*e1(*x*,*t*): ionic current density through the electrolyte in the negative electrode, A m–2

*i*e2(*x*,*t*): ionic current density through the electrolyte in the positive electrode, A m–2

*i*e3(*x*,*t*): ionic current density through the electrolyte in the separator, A m–2

: Faradaic current density in the negative electrode is defined as , A m–2

: interfacial current density in the negative electrode is defined as , A m–2

: interfacial current density in the positive electrode is defined as , A m–2

*i*s1(*x*,*t*): electronic current density through the solid phase in the negative electrode, A m–2

*i*s2(*x*,*t*): electronic current density through the solid phase in the positive electrode, A m–2

*j*1(*x*,*t*): reaction flux between the electrolyte and the negative electrode particle, mol m–2 s–1

*j*2(*x*,*t*): reaction flux between the electrolyte and the positive electrode particle, mol m–2 s–1

: complex phasor for *j*1(*x*,*t*), mol m–2 s–1

*j*F1(*x*,*t*): molar flux of Li-ions de-intercalating out of the solid phase in the negative electrode, mol m–2 s–1

: complex phasor for *j*F1(*x*,*t*), mol m–2 s–1

*j*dl1(*x*,*t*): electrochemical double-layer flux in the negative electrode, mol m–2 s–1

: complex phasor for *j*dl1(*x*,*t*), mol m–2 s–1

*k*1: rate constant for the negative electrode electrochemical reaction, mol/(m2 s)/(mol/m3)1+*α*a1

*k*2: rate constant for the positive electrode electrochemical reaction, mol/(m2 s)/(mol/m3)1+*α*a2

*L*1: thickness of the negative electrode, m

*L*2: thickness of the positive electrode, m

*L*3: thickness of the separator, m

*L*4: thickness of the cell is defined as , m

*r*: radial position across a spherical particle, m

*R*: universal gas constant, 8.314 J mol–1 K–1

*R*ct1: polarization resistance for charge transfer reaction at the solid/electrolyte interface in the negative electrode at the particle scale is defined as , Ω m2

*R*CT1: polarization resistance for charge transfer reaction at the solid/electrolyte interface in the negative electrode at the electrode scale, Ω m2

*R*d1: polarization resistance for solid diffusion in the negative electrode at the particle scale, Ω m2

*R*De1: polarization resistance for electrolyte diffusion in the negative electrode, Ω m2

*R*De3: polarization resistance for electrolyte diffusion in the separator, Ω m2

*R*diff1: solid phase diffusion resistance in the negative electrode particle is defined as , Ω m2

*R*Ds1: polarization resistance for solid diffusion in the negative electrode at the electrode scale, Ω m2

*R*Ds2: polarization resistance for solid diffusion in the positive electrode at the electrode scale, Ω m2

*R*O3: ohmic resistance in the separator is defined as , Ω m2

*r*s1: particle radius in the negative electrode, m

*r*s2: particle radius in the positive electrode, m

*R*sei1: polarization resistance for charge transport through sei film in the negative electrode at the particle scale is defined as , Ω m2

*R*SEI1: polarization resistance for charge transport through SEI film in the negative electrode at the electrode scale, Ω m2

*R*sei2: polarization resistance for charge transport through sei film in the positive electrode at the particle scale, Ω m2

*s*: frequency variable is defined as , s–1

: dimensionless frequency variable in the negative electrode is defined as

: dimensionless frequency variable in the positive electrode is defined as

: dimensionless frequency variable in the separator is defined as

SOC0: initial SOC of the electrode

*SZ*f: the standard derivation of *Zf*,*i* over *M* sampling frequency points (*M* = 38), Ω m2

*SZ*%: the relative sensitivity of the parameter *X* to the solid/electrolyte diffusion processes

*t*: time, s

*T*: ambient temperature, 298.15 K

*t*0 +: transference number of Li+ with respect to the velocity of solvent

*U*ocp1(*c*ss1): open-circuit potential of the negative electrode, V

*U*ocp2(*c*ss2): open-circuit potential of the positive electrode, V

*x*: position across cell, m

: dimensionless thickness of the negative electrode is defined as

: dimensionless thickness of the positive electrode is defined as

: dimensionless thickness of the separator is defined as

*Y*s1(*s*): transfer function for the solid phase diffusion in a single particle is defined as

*Z*1(*s*): negative electrode impedance for Model DFN, Ω m2

*Z*2(*s*): positive electrode impedance for Model DFN, Ω m2

*Z*3(*s*): separator impedance for Model DFN, Ω m2

*Z*4(*s*): full cell impedance for Model DFN, Ω m2

*Z*A#1(*s*): negative electrode impedance for Model A, Ω m2

*Z*B#1(*s*): negative electrode impedance for Model B, Ω m2

*Z*C#1(*s*): negative electrode impedance for Model C, Ω m2

*z*d1(*s*): the complex impedance for the solid diffusion in the negative electrode at the particle scale is defined as , Ω m2

*Z*D1: full diffusion impedance at the electrode scale in the negative electrode is defined as , Ω m2

*Z*De1: electrolyte diffusion impedance in the negative electrode is defined as , Ω m2

*Z*Ds1: solid diffusion impedance in the negative electrode at the electrode scale is defined as , Ω m2

*Z*D#1(*s*): negative electrode impedance for Model D, Ω m2

*Z*E#1(*s*): negative electrode impedance for Model E, Ω m2

*Z*F#1(*s*): negative electrode impedance for Model F, Ω m2

*z*F1(*s*): the complex impedance for the Faradaic behavior in the negative electrode at the particle scale is defined as , Ω m2

*Zf*,*i*: the ith simulating result of the solid/electrolyte diffusion impedance, Ω m2

: the average value of *Zf*,*i* at frequency *f* over *i* = 1–*N* (*N* = 5) times simulating results

*z*int1(*s*): the complex impedance for the whole process with sei film in the negative electrode at the particle scale is defined as , Ω m2

*z*int2(*s*): the complex impedance for the whole process with sei film in the positive electrode at the particle scale is defined as , Ω m2

#### S3.3. Greek symbols

*τ*ct1: characteristic time constant for charge transfer reaction at the solid/electrolyte interface in the negative electrode at the particle scale is defined as , s

*τ*CT1: characteristic time constant for charge transfer reaction at the solid/electrolyte interface in the negative electrode at the electrode scale, s

*τ*d1: characteristic time constant for solid diffusion in the negative electrode at the particle scale is defined as , s

*τ*De1: characteristic time constant for electrolyte diffusion in the negative electrode at the electrode scale, s

*τ*De3: characteristic time constant for electrolyte diffusion in the separator, s

*τ*Ds1: characteristic time constant for solid diffusion in the negative electrode at the electrode scale, s

*τ*sei1: characteristic time constant for charge transport through sei film in the negative electrode at the particle scale is defined as , s

*τ*SEI1: characteristic time constant for charge transport through SEI film in the negative electrode at the electrode scale, s

*α*a1: anodic charge transfer coefficient in the negative electrode

*α*a2: anodic charge transfer coefficient in the positive electrode

*α*c1: cathodic charge transfer coefficient in the negative electrode

*α*c2: cathodic charge transfer coefficient in the positive electrode

*δ*sei1: sei thickness in the negative electrode, m

*ε*e1: electrolyte phase volume fraction(porosity) in the negative electrode

*ε*e2: electrolyte phase volume fraction(porosity) in the positive electrode

*ε*e3: electrolyte phase volume fraction(porosity) in the separator

*ε*f1: conductive filler volume fraction in the negative electrode

*ε*f2: conductive filler volume fraction in the positive electrode

*ε*s1: active particles volume fraction in the negative electrode

*ε*s2: active particles volume fraction in the positive electrode

*ε*sei1: sei permittivity in the negative electrode, F m–1

*θ*1,0%: stoichiometry at 0% SOC in the negative electrode

*θ*2,0%: stoichiometry at 0% SOC in the positive electrode

*θ*1,100%: stoichiometry at 100% SOC in the negative electrode

*θ*2,100%: stoichiometry at 100% SOC in the positive electrode

*σ*: electronic conductivity of solid matrix, S m–1

*σ*eff1: effective solid conductivity in the negative electrode is defined as , S m–1

*σ*eff2: effective solid conductivity in the positive electrode is defined as , S m–1

*κ*: ionic conductivity of electrolyte, S m–1

*κ*eff1: effective electrolyte conductivity in the negative electrode is defined as , S m–1

*κ*eff2: effective electrolyte conductivity in the positive electrode is defined as , S m–1

*κ*eff3: effective electrolyte conductivity in the separator is defined as , S m–1

*κ*D,eff1: effective diffusional conductivity in the negative electrode is defined as , A m–1

*κ*D,eff2: effective diffusional conductivity in the positive electrode is defined as , A m–1

*ρ*sei1: sei resistivity in the negative electrode, Ω m

*φ*e1(*x*,*t*): electrolyte phase potential in the negative electrode, V

*φ*e2(*x*,*t*): electrolyte phase potential in the positive electrode, V

*φ*e3(*x*,*t*): electrolyte phase potential in the separator, V

: complex phasor for *φ*e(*x*,*t*), V

: dimensionless electrolyte potential in the separator is defined as

*φ*s1(*x*,*t*): solid phase potential in the negative electrode, V

*φ*s2(*x*,*t*): solid phase potential in the positive electrode, V

: debiased version of *φ*s(*x*,*t*) is defined as , V

: complex phasor for , V

*φ*s−e1(*x*,*t*): potential difference between solid and electrolyte phases in the negative electrode is defined as , V

*φ*s−e2(*x*,*t*): potential difference between solid and electrolyte phases in the positive electrode is defined as , V

: debiased version of *φ*s−e(*x*,*t*) is defined as , V

: complex phasor for , V

: dimensionless potential difference in the negative electrode is defined as

: dimensionless potential difference in the positive electrode is defined as

*φ*sf1(*x*,*t*): potential in the film close to the solid phase in the negative electrode, V

: complex phasor for *φ*sf1(*x*,*t*), V

*η*1(*x*,*t*): overpotential in the negative electrode, V

*η*2(*x*,*t*): overpotential in the positive electrode, V

: concentration flux at the negative electrode/separator interface, mol m–2 s–1

: concentration flux at the positive electrode/separator interface, mol m–2 s–1

: phasor form of , mol m–2 s–1

: phasor form of , mol m–2 s–1

ΘI1(*s*): the ratio of the electrolyte-related diffusion resistance to *z*int1 in the negative electrode is defined as

ΘI2(*s*): the ratio of the electrolyte-related diffusion resistance to *z*int2 in the positive electrode is defined as

ΘII1(*s*): the ratio of the whole electrode-related ohmic resistance to *z*int1 in the negative electrode is defined as

ΘII2(*s*): the ratio of the whole electrode-related ohmic resistance to *z*int2 in the positive electrode is defined as

ΘIII3: dimensionless parameter in the separator is defined as

: relative difference for Model A is defined as , n=1,2,3,4

: relative difference for Model B is defined as , n=1,2,4

: relative difference for Model C is defined as , n=1,2,4

#### S3.4. Subscripts

0: initial value or reference parameter value at reference temperature

1: negative electrode

2: positive electrode

3: separator

4: full cell

a: anodic

c: cathodic

ct: charge-transfer at the particle scale

CT: charge-transfer at the electrode scale

d: solid diffusion at the particle scale

De: electrolyte diffusion at the electrode scale

dl: double-layer at the particle scale

DL: double-layer at the electrode scale

Ds: solid diffusion at the electrode scale

e: electrolyte

eff: effective value

f: conductive filler

F: Faradaic process

max: maximum value

s: solid matrix

sei: solid electrolyte interlay at the particle scale

SEI: solid electrolyte interlay at the electrode scale

ss: solid surface

#### S3.5. Decorations

tilde “~”: debiased variable

point “·”: plural identities related to sine quantities

dash “−”: dimensionless variable