

Electro-Chemo-Mechanical Modelling of Structural Battery Full Cells

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Electro-chemo-mechanical modelling of structural battery composite full cells

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Structural battery composites are multifunctional materials capable of storing electrochemical energy and carry mechanical load at the same time. In this study, we focus on the laminated structural battery design developed by Asp and co-workers, which utilises multifunctional carbon fibres as both active material and mechanical reinforcement in the negative electrode. The positive electrode consists of active lithium iron phosphate particles adhered to an aluminium foil. Building upon previous research, we develop a fully coupled numerical multiphysics model to simulate the charge–discharge processes of the structural battery full cell. The model includes non-linear reaction kinetics, pertinent to the Butler–Volmer relation. Furthermore, we employ a simplified continuum representation of the porous positive electrode, enabling simulations at the battery cell level. Available experimental data for material parameters is utilised when possible, while the remaining parameters are obtained from calibration against experimental charge–discharge voltage profiles at two different rates. Results show that the presented model captures the general trend of the experimental voltage profiles for a range of charge rates. Through this work, we aim to provide insights for future structural battery design efforts.

Structural battery composites are among the group of multifunctional materials that offer the storage of electrochemical energy in the mechanical load path of a structure. A recent structural battery developed by Asp and co-workers uses carbon fibres both as a negative electrode and as mechanical reinforcement^{1,2}. The commerically available positive electrode consists of active lithium iron phosphate (LiFePO₄, LFP) particles together with a conductive binder material adhered to an aluminium foil. The battery composite is completed by embedding the positive and negative electrodes together with a separator, in a two-phase matrix called structural battery electrolyte (SBE)³. The solid phase of the SBE bonds the constituents, enabling mechanical load transfer between the fibres. The liquid electrolyte phase, on the other hand, allows for lithium-ion transfer between the electrodes.

Carlstedt et al.⁴ proposed the first fully coupled numerical multiphysics model of a structural battery composite in a negative half-cell representation, i.e., carbon fibres vs. lithium metal. The model included features such as chemo-mechanical coupling, known as stress-assisted diffusion or piezoelectrochemical transducer (PECT) effect, relevant for the structural battery application⁵. Furthermore, Carlstedt et al.⁶ assessed the model performance by simulating charge/discharge processes and comparing them against experiments. Three different models were evaluated against the experiment, each having a unique formulation of the chemical potential. Reaction kinetics on the electrode/electrolyte interfaces were modelled using a linearised relation of the Butler–Volmer type. This approximation is accurate for low galvanostatic charge/discharge rates, resulting in low overpotentials, but is less precise when higher rates are introduced⁷. Results show that regardless of the chemical potential formulation, the model struggles to capture discontinuities in the cell potential curve as the current is changed from charge to rest. To date, no attempt has been made to model the coupled electro-chemo-mechanical processes in a structural positive electrode halfcell nor in a structural battery full cell.

The porous LFP-based positive electrode comes in various particle sizes ranging from nano to microsized particles. As discussed by Grazioli et al.⁸, multiphysics phenomena take place at different length scales. However, it is not computationally feasible to resolve nano-sized LFP particles if simulations are to be carried out at the battery cell level. To circumvent this issue, Newman et al.^{9,10} developed models of porous electrodes in a macroscopic, averaged sense. More recent works on homogenisation of composite electrodes for conventional batteries have been carried out by Salvadori et al.¹¹, outlining the micro–macro scale transition in a coupled, thermo-dynamically consistent way.

In this paper, we model the charge–discharge process of the laminated structural battery full cell using a simplified, continuum (macroscale) representation of the positive electrode. Starting with a simplistic model, we

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Fig. 1 | **Galvanostatic charge-discharge profiles. a** Experimental and calibrated voltage profiles at C/20 for Cell 1. b Experimental and calibrated voltage profiles at C/10 for Cell 1. c Applied current profile for the charge–rest–discharge cycle at C/20.

d Applied current profile for the charge–rest–discharge cycle at C/10. The shaded grey areas in **a** and **b** indicate the experimental data spread.



can gain insight into the level of complexity required for a representative description of the positive electrode based on our experimental observations. This understanding allows us to incrementally introduce additional complexity in a structured manner. Moreover, recent advancements in state-of-the-art positive electrode design involve carbon fibres coated with active material, altering the geometry of the electrode¹². The evolving design further emphasises the need for a flexible modelling approach. Furthermore, we adopt non-linear reaction kinetics pertinent to the Butler–Volmer relation adapted to the proposed modelling framework and present chemo-mechanical coupling on the electrodes and mechanical considerations for all domains. Finally, the model is calibrated against experimental charge/discharge cycles at two different charge rates for a single cell and validated for an independent case, i.e. a different cell and charge rate.

Results

Model and calibration outcomes

Figure 1 displays both the experimental and simulated cell voltage profiles for the optimised material parameters θ^* along with the corresponding applied current profiles over time. In Fig. 1a and b, it is evident that the calibrated model captures the general trend of the charge-rest-discharge voltage profiles. The experimental voltage profiles exhibit a pronounced non-linear evolution of the potential during charge and discharge, which seemingly increases with decreasing charge rate. The model is currently not able to fully capture the non-linear evolution but rather produces a fairly linear cell potential increase/decrease during charge and discharge. The trend towards a more linear potential evolution for increasing charge rates is also observed for the validation case of the second cell shown in Fig. 2. Here, the cell potential slope exhibits a more linear behaviour during charge and discharge, leading to a more accurate prediction of the model.

Furthermore, the lithium concentration field during charge, rest and discharge, respectively, simulated at C/10 is shown in Fig. 3. During charge,

respectively, simulated at C/10 is shown in Fig. 5. During ch

the carbon fibres closest to the separator have higher concentrations of lithium, and the carbon fibres farther away from the separator contain less lithium. At cell rest (zero current), a pronounced lithium concentration gradient is observed in the carbon fibres and electrolyte domains, which will relax with time. During discharge, the opposite situation occurs, where the carbon fibres closest to the separator are emptied first, and the fibres farther away contain higher concentrations of lithium.

The magnitude of the conductivity and transference number are in line with what is reported by Cattaruzza et al. and Pipertzis et al.^{13,14}. Both the conductivity and transference number relate to the corresponding mobility coefficients in the SBE, which in turn are related to diffusion coefficients for comparison.

The resulting diffusion coefficients, presented in Table 1 are of comparable magnitude. The dominance of SBE properties during cell relaxation can be related to the fact that, at zero current, the electric potential gradient becomes zero, eliminating migration as a transport mechanism. Considering that the separator thickness is ~60 times greater than the diameter of a carbon fibre, and the diffusion coefficients are of a similar order of magnitude, it is expected that the limiting transport properties are t_+ and κ_{SBE} . Figure 4 shows the evaluated sensitivity modes, where each parameter is perturbed by 5%. Figure 4a shows the sensitivity of the conductivity κ_{SBE} and transference number t^+ in the SBE. Perturbation of these parameters causes a change of slope in the cell potential during charge and discharge, as well as affects the rate of lithium redistribution during cell rest. The fact that the cell potential is significantly affected during cell rest further indicates that the bottleneck related to lithium redistribution at zero current over time in a structural battery full cell, is the transport properties t_+ and κ_{SBE} .

As the current switches from charge to rest, the drop in cell potential is overestimated both for the calibration and validation voltage profiles. This is mainly due to the interface property $i_{0,P}$ and the related parameters q_0-q_3 shown in Fig. 4b. Variation in these parameters causes a change in the slope



Fig. 3 | Lithium concentration distributions. Lithium concentration distribution c_{Liv} at different times of the charge–rest–discharge cycle simulated at C/10.

Table 1 | Diffusion coefficients pertinent to the specific domains

Parameter	Description	Value
D _{Li,CF}	Average diffusion coefficient in the carbon fibres	$1.12 \times 10^{-12} m^2 s^{-1}$
D _{Li}	Diffusion coefficient of lithium ions in the SBE	$2.89\times 10^{-12}m^2s^{-1}$
D _X	Diffusion coefficient of X anions in the SBE	$4.96\times 10^{-12}m^2s^{-1}$
Ē_⊥i	Diffusion coefficient of lithium ions in the separator	$1.9 \times 10^{-12} m^2 s^{-1}$
Ū _X	Diffusion coefficient of X anions in the separator	$3.27 \times 10^{-12} m^2 s^{-1}$
D _{Li,P}	Effective diffusion coefficient in the positive electrode	$1.0 \times 10^{-11} \mathrm{m^2 s^{-1}}$

of the cell potential, as well as the discontinuity obtained when the current is switched from charge to rest and from rest to discharge.

The sensitivity analysis further shows that $k_{\rm CF}$ primarily affects the slope of the cell potential with respect to time in a linear manner, illustrated in Fig. 4c. In contrast, the maximum concentrations of lithium in the carbon

fibres and the positive electrode, shown in Fig. 4d, impact the potential response during charging in a non-linear sense. Both the solubility parameter and the maximum concentration are incorporated into the expression for the chemical potential of lithium in the carbon fibres. As expected, perturbing the solubility parameter results in a linear change in the cell potential, while perturbing the maximum concentration induces a non-linear change. This is because the chemical potential, $\mu_{\rm Li}$, varies linearly with $k_{\rm CF}$ but exhibits non-linear dependence on $\tilde{c}_{\rm CF}$.

Furthermore, the effective diffusion coefficient, solubility parameter, and electrical conductivity associated with the homogenised positive electrode, $D_{\text{Li},\text{P}}$, k_{P} , and κ_{P} , respectively, show negligible impact on the cell potential for a 5% perturbation and are therefore not presented in a graph. All numerical values for the model parameters considered in the analysis are stated in Table 2 along with a list of symbols in Table 3.

Discussion

In this study, we present a first-ever model of a structural battery full cell. Measured data for parameters are used when available, while the remaining parameters are determined via calibration against experimental charge–rest–discharge voltage profiles. The influence of the individual



Fig. 4 | **Sensitivity analysis.** Sensitivity modes computed using Eq. (82) subjected to a 5% perturbation, vs time. The black dashed lines separate the different stages of the applied current profile, signifying the charge, rest and discharge processes in Fig. 2b. Sensitivities are illustrated for: **a** the conductivity κ_{SBE} and transference number t^+ .



b the parameters related to the exchange current density in the positive electrode, $i_{0,P}$. **c** solubility parameter k_{CF} related to the chemical potentials in the carbon fibres, and **d** the maximum concentrations in the positive electrode, \tilde{c}_{P} , and carbon fibres, \tilde{c}_{CF} .

Table 2 | Table of material parameters

Parameters	Description	Value	Source
EL	Longitudinal carbon fibre modulus	294(1-0.12c _{Li}) GPa	15
E _T	Transverse carbon fibre modulus	21.8(1 + 1.07c _{Li}) GPa	15
G _{LT}	Longitudinal-transverse shear modulus	12.5 GPa	Assumed
ν _{TT}	Transverse-transverse carbon fibre Poisson ratio	0.2	Assumed
V _{LT}	Longitudinal–transverse carbon fibre Poisson ratio	0.22	Assumed
a _T	Transverse expansion coefficient	0.066	15
aL	Longitudinal expansion coefficient	0.0085	6
E	SBE Young's modulus	0.7 GPa	18
v	SBE Poisson ratio	0.37	Assumed
κ _N	Carbon fibre electric conductivity	71,429 S m ⁻¹	25
КР	Positive electrode electric conductivity	$0.0211\mathrm{S}\mathrm{m}^{-1}$	Calibrated
\widetilde{c}_{CF}	Maximum Li concentration in carbon fibres	15,609 mol m ⁻³	Calibrated
€ _P	Maximum Li concentration in positive electrode	10,373 mol m ⁻³	Calibrated
c ⁰ _{CF}	Initial Li concentration in carbon fibres	$0.99\widetilde{c}_{\rm CF}~{ m mol}~{ m m}^{-3}$	Assumed
c _P ⁰	Initial Li concentration in positive electrode	$0.01\widetilde{c}_{P} \text{ mol } m^{-3}$	Assumed
C _{ref}	Reference concentration of Li in electrolyte	1000 mol m ⁻³	26
ε	Electric permittivity in electrolyte	$8.854 \times 10^{-11} F m^{-1}$	Assumed
٤p	Electric permittivity in the positive electrode	$8.854 \times 10^{-11} F m^{-1}$	Assumed
ε _N	Electric permittivity in the carbon fibres	$8.854 \times 10^{-11} F m^{-1}$	Assumed
D _{Li,CF}	Diffusion coefficient in carbon fibres	See Eq. (67)	21
i _{0,N}	Exchange current density function in carbon fibres	See Eq. (69)	21
D _{Li,P}	Mobility coefficient in positive electrode	$1 \times 10^{-11} m^2 s^{-1}$	Calibrated
<i>q</i> ₀	Exchange current density parameter	0.0168	Calibrated
<i>q</i> ₁	Exchange current density parameter	0	Calibrated
<i>q</i> ₂	Exchange current density parameter	-0.0162	Calibrated
<i>q</i> ₃	Exchange current density parameter	3.54×10^{-4}	Calibrated

Table 2 (continued) | Table of material parameters

Parameters	Description	Value	Source
<i>t</i> +	Transference number	0.3684	Calibrated
KSBE	SBE conductivity	$0.15Sm^{-1}$	Calibrated
$\mu_{\rm CF}^0$	Reference chemical potential in carbon fibres	$6.7805 \times 10^4 J mol^{-1}$	6
$\mu_{\rm P}^0$	Reference chemical potential in positive electrode	$-2.9659 \times 10^{5} J mol^{-1}$	Calibrated
$\mu^0_{\rm Li}$	Reference chemical potential of Li in SBE	0 J mol ⁻¹	6
μ_X^0	Reference chemical potential of X in SBE	0 J mol ⁻¹	6
k _{CF}	Solubility parameter in carbon fibres	-39.8035	Calibrated
<i>k</i> _P	Solubility parameter in positive electrode	5.0487	Calibrated
Т	Temperature	293.15 K	Assumed

parameters on the predicted, i.e. simulated, cell voltage profile is assessed using sensitivity analysis. The model effectively captures the general trends of experimental charge-rest-discharge profiles across various charge rates. The calibration of model parameters against experimental data has demonstrated the model's capability to predict voltage profiles with reasonable accuracy, although some discrepancies in non-linear transient behaviour were observed. A simplified representation of the positive electrode is employed where underlying physical phenomena are considered for the homogenised material representation. For instance, redox reactions on the LiFePo₄ particle interfaces and the related particle swelling due to Li-insertion are disregarded. A more sophisticated model for the homogenised positive electrode is required to accurately predict charge-rest-discharge profiles of structural battery composites. Furthermore, introducing more physics also means that more experimental material-level tests can be employed to populate the model, thus decreasing the amount of parameters needed for calibration.

Using the proposed method in combination with sensitivity analysis, we gain insight into which material property is limiting during charge, rest and discharge. In this way, we use the modelling results to guide further development of structural battery composites. For instance, it was found that the transport properties within the SBE play a crucial role in the transient response, suggesting that future research should prioritise enhancing these properties to improve overall battery performance. This work enhances the fundamental understanding of structural battery composites and provides a foundation for future research in energy storage technologies. Finally, we note that the modelling framework established here is versatile and can be adapted also for conventional lithium-ion batteries, thereby broadening its applicability and impact in the field of energy storage solutions.

Methods

Preliminaries

The analysis considers a synthetical microscale volume element $\Omega = \Omega_{CF} \cup \Omega_{SBE} \cup \Omega_{SEP} \cup \Omega_P$ subdivided into several domains, shown in Fig. 5. Ω_{CF} represents the carbon fibre domain in the negative electrode, described by the union of carbon fibres in the volume

Table 3 | List of symbols

Quantity	Description Unit	
σ	Mechanical stress tensor [Pa]	
e	Mechanical strain tensor [-]	
E	Elasticity tensor [Pa]	
а	Li-induced expansion tensor [-]	
u	Displacement vector [m]	
Ca	Concentration of species a [mol m ⁻³]	
j a	Flux of species α [mol m ⁻² s ⁻¹	
μ_a	Chemical potential of species a [J mol ⁻¹]	
D	Diffusion coefficient	$[m^2 s^{-1}]$
ηα	Mobility coefficient of species <i>a</i> [m ² mol s ⁻	
R	Ideal gas constant	[J K ⁻¹ mol ⁻¹]
d	Electric flux density vector [C m ⁻²]	
i	Electric flux	[A m ⁻²]
φ	Local electric potential	[V]
Φ	Electrode potential	[V]
к	Electric conductivity	[S m ⁻¹]
ε	Electric permittivity	[F m ⁻¹]
i ₀	Exchange current density	[A m ⁻²]
η_N	Interface overpotential at the negative electrode	[V]
$\eta_{\rm P}$	Interface overpotential at the positive electrode	[V]
Za	Charge number of species a	[-]
F	Faraday's Constant	[C mol ⁻¹]
ψ	Helmholtz free energy	[J]
t	Time	[S]
n	Unit normal	[-]
Ω	Domain	[-]
Г	Boundary	[-]

element. The carbon fibres are embedded in a structural battery electrolyte matrix denoted Ω_{SBE} , where $\Omega_N = \Omega_{CF} \cup \Omega_{SBE}$ is referred to as the negative electrode. Redox reactions occur on the carbon fibre/SBE interfaces, $\Gamma_{\rm CF} = \partial \Omega_{\rm CF} \cap \partial \Omega_{\rm SBE}$. To prevent short circuits, the positive and negative electrodes are separated by a porous SBE-impregnated glass fibre separator, Ω_{SEP} . The interface between the negative electrode and the separator is denoted $\Gamma_N^{int} = \partial \Omega_N \cap \partial \Omega_{SEP}$. For convenience, we define the electrolyte domain (ED) where ionic transport takes place as $\Omega_{ED} = \Omega_{SEP} \cup \Omega_{SBE}$. Ω_P denotes the homogenised positive electrode domain, where $\Gamma_{\rm p}^{\rm int} = \partial \Omega_{\rm SEP} \cap \partial \Omega_{\rm p}$ describes the interface between the positive electrode and separator domains. The cell thickness, h_{cell} , is roughly 390 µm, whereas the negative electrode has a thickness of $h_{\rm N} = 50 \,\mu{\rm m}$. The thickness of the Whatman GF/A separator is approximately $h_{\text{SEP}} = 260 \,\mu\text{m}$, while the positive electrode has a thickness of $h_{\rm P} = 80 \,\mu {\rm m}^{1.6}$. The positive electrode is conceptualised as a continuum, implying that the underlying structure is not resolved. It is acknowledged that redox reactions occur on each interface of the active LFP particles distributed at various positions inside $\Omega_{\rm P}$. To simplify the modelling approach, the redox reactions in the positive electrode are modelled on the Γ_P^{int} interface. Through this simplification, the interface kinetics are clearly structural, and will vary with, e.g., the thickness of the positive electrode. Moreover, we assume that both the positive and negative electrodes function as ideal conductors. This implies that all carbon fibres are electrically connected to a current collector, establishing the global potential of the negative electrode, denoted $\Phi_{\rm N}$ inside $\Omega_{\rm CF}$. The same rationale applies to the positive electrode characterised by the global potential $\Phi_{\rm P}$ inside $\Omega_{\rm P}$.

In addition to the interfaces already discussed, Fig. 5 illustrates an assumed periodic structure in the 2-direction. The right side $\Gamma^+ = \Gamma_{\rm N}^+ \cup \Gamma_{\rm SEP}^+ \cup \Gamma_{\rm P}^+$ has a corresponding mirror side $\Gamma^- = \Gamma_{\rm N}^- \cup \Gamma_{\rm SEP}^- \cup \Gamma_{\rm P}^-$. For the subsequent introduction of periodic boundary conditions, we introduce the mapping of points $\varphi_{\rm PER}$: $\Gamma^+ \to \Gamma^-$ such that $f^-(\mathbf{x}^+) = f(\varphi_{\rm PER}(\mathbf{x}^+))$ and $f^+(\mathbf{x}^+) = f(\mathbf{x}^+)$ for any function $f(\mathbf{x})$ on $\Gamma^+ \cup \Gamma^-$.

Balance equations

In the following, we formulate the boundary value problem for the domains shown in Fig. 5. The balance of linear momentum and relevant boundary conditions applicable to the negative electrode domain is stated as

$$-\boldsymbol{\sigma} \cdot \boldsymbol{\nabla} = \boldsymbol{0} \quad \text{in} \quad \boldsymbol{\Omega}_{N} \cup \boldsymbol{\Omega}_{SEP} \cup \boldsymbol{\Omega}_{P}, \tag{1}$$

$$\boldsymbol{u}^+ - \boldsymbol{u}^- = \boldsymbol{0}, [\boldsymbol{\sigma}^+ - \boldsymbol{\sigma}^-] \cdot \boldsymbol{n}^+ = \boldsymbol{0} \quad \text{on} \quad \Gamma_{\mathrm{N}}^+ \cup \Gamma_{\mathrm{SEP}}^+ \cup \Gamma_{\mathrm{P}}^+, \quad (2)$$



Fig. 5 | Structural battery model description. Schematic two-dimensional illustration of the structural battery domains and boundaries.

q

$$\boldsymbol{\sigma} \cdot \boldsymbol{n} = \boldsymbol{0} \quad \text{on} \quad \boldsymbol{\Gamma}_{\mathrm{N}}^{\mathrm{ext}} \cup \boldsymbol{\Gamma}_{\mathrm{P}}^{\mathrm{ext}}, \tag{3}$$

where σ is the Cauchy stress tensor, u is the displacement field and n is the (from Ω_N) outwards facing unit normal.

The ionic transport of lithium ions and the accompanying anions, here denoted X, inside $\Omega_{\rm ED}$ is governed by the conservation of mass and Gauss law as follows:

$$\dot{c}_{\alpha} + \boldsymbol{j}_{\alpha} \cdot \nabla = 0$$
 in $\Omega_{\rm ED}$, $\alpha = {\rm Li}, {\rm X},$ (4)

$$F[c_{\rm Li} - c_X] - \boldsymbol{d} \cdot \nabla = 0 \quad \text{in} \quad \Omega_{\rm ED}, \tag{5}$$

$$\mu_{\alpha}^{+} - \mu_{\alpha}^{-} = 0, [\boldsymbol{j}_{\alpha}^{+} - \boldsymbol{j}_{\alpha}^{-}] \cdot \boldsymbol{n}^{+} = 0 \quad \text{on} \quad \Gamma_{\text{SEP}}^{+} \cup \Gamma_{\text{N}}^{+}, \quad \alpha = \text{Li}, \text{X}, \quad (6)$$

$$\Gamma^{+} - \varphi^{-} = 0, [d^{+} - d^{-}] \cdot n^{+} = 0 \quad \text{on} \quad \Gamma^{+}_{\text{SEP}} \cup \Gamma^{+}_{\text{N}},$$
(7)

$$\boldsymbol{j}_{\mathrm{Li}} \cdot \boldsymbol{n} = -\boldsymbol{j}_{\mathrm{P}}, \boldsymbol{d} \cdot \boldsymbol{n} = -\boldsymbol{d}_{\mathrm{P}} \quad \text{on} \quad \boldsymbol{\Gamma}_{\mathrm{P}}^{\mathrm{int}},$$
 (8)

$$\boldsymbol{j}_{\mathrm{Li}} \cdot \boldsymbol{n} = \boldsymbol{j}_{\mathrm{N}}, \boldsymbol{d} \cdot \boldsymbol{n} = \boldsymbol{d}_{\mathrm{N}}, \quad \text{on} \quad \Gamma_{\mathrm{CF}}$$
(9)

$$\boldsymbol{j}_{\mathrm{Li}} \cdot \boldsymbol{n} = 0, \boldsymbol{d} \cdot \boldsymbol{n} = 0 \quad \text{on} \quad \Gamma_{\mathrm{N}}^{\mathrm{ext}},$$
 (10)

$$\boldsymbol{j}_{X} \cdot \boldsymbol{n} = 0 \quad \text{on} \quad \boldsymbol{\Gamma}_{\mathrm{P}}^{\mathrm{int}} \cup \boldsymbol{\Gamma}_{\mathrm{N}}^{\mathrm{ext}} \cup \boldsymbol{\Gamma}_{\mathrm{CF}}. \tag{11}$$

where c_{α} and j_{α} denote the concentration and flux, respectively, of lithium ions ($\alpha = \text{Li}$) and the accompanying anion ($\alpha = X$). μ_{α} denotes the chemical potential for the ions ($\alpha = \text{Li}$, X), d is the electric flux density, F is Faraday's constant and φ is the electric potential. Here, n denotes the outwards pointing normal (from Ω_{ED}). Hence, during charging, j_P denotes the flux of Li-ions entering the electrolyte from the positive electrode, and j_N denotes the flux of Li-ions moving into the negative electrode. Similarly, d_P and d_N denote the electric flux density moving from the positive and to the negative electrode, respectively.

Finally, the transport of charge-neutral lithium inside the electrodes is governed by mass conservation as

$$\dot{c}_{\mathrm{Li}} + \boldsymbol{j}_{\mathrm{Li}} \cdot \boldsymbol{\nabla} = 0 \quad \text{in} \quad \boldsymbol{\Omega}_{\mathrm{P}} \cup \boldsymbol{\Omega}_{\mathrm{CF}}, \tag{12}$$

$$\mu_{\rm Li}^{+} - \mu_{\rm Li}^{-} = 0, \left[\boldsymbol{j}_{\rm Li}^{+} - \boldsymbol{j}_{\rm Li}^{-} \right] \cdot \boldsymbol{n}^{+} = 0 \quad \text{on} \quad \Gamma_{\rm P}^{+}, \tag{13}$$

$$\boldsymbol{j}_{\mathrm{Li}} \cdot \boldsymbol{n} = \boldsymbol{j}_{\mathrm{P}} \quad \text{on} \quad \boldsymbol{\Gamma}_{\mathrm{P}}^{\mathrm{int}}, \tag{14}$$

$$\boldsymbol{j}_{\mathrm{Li}} \cdot \boldsymbol{n} = -\boldsymbol{j}_{\mathrm{N}} \quad \text{on} \quad \Gamma_{\mathrm{CF}}$$
 (15)

$$\boldsymbol{j}_{\mathrm{Li}} \cdot \boldsymbol{n} = 0 \quad \text{on} \quad \Gamma_{\mathrm{P}}^{\mathrm{ext}},$$
 (16)

where c_{Li} and j_{Li} denote the concentration and flux, respectively, of neutral lithium atoms. Here, n is the outwards pointing unit normal (from $\Omega_{\text{CF}}/\Omega_{\text{P}}$). Hence, the conditions in Eqs. (13) and (14) combined with Eqs. (8) and (9) pertain to mass conservation over the interfaces $\Gamma_{\text{P}}^{\text{int}}$ and Γ_{CF} . Across the interfaces $\Gamma_{\text{p}}^{\text{int}}$ and Γ_{CF} , we introduce the jump operators

$$\llbracket \mu_{\mathrm{Li}} \rrbracket := \mu_{\mathrm{Li}}^+ - \mu_{\mathrm{Li}}^-, \quad \mu_{\mathrm{Li}}^\pm := \lim_{\epsilon \downarrow 0} \mu_{\mathrm{Li}}(\mathbf{x} \pm \epsilon \mathbf{n}_{\Gamma})$$
(17)

for the normal n_{Γ} associated with the convention for $j_{\rm P}$, $j_{\rm N}$. Hence, n_{Γ} points from $\Omega_{\rm P}$ to $\Omega_{\rm SEP}$ and from $\Omega_{\rm SBE}$ to $\Omega_{\rm CF}$.

When solving the pertinent initial boundary value problem, we introduce the spatially constant electric potentials Φ_N in Ω_{CF} and Φ_P in Ω_P . The total current density over the positive electrode is evaluated at the

interface Γ_P^{int} as

$$I_{\rm app} = F \int_{\Gamma_{\rm p}^{\rm int}} j_{\rm P} \, \mathrm{d}\,\Gamma,\tag{18}$$

where I_{app} is the prescribed current for a galvanostatic (current control) process. Furthermore, the initial, stress-free reference state is given at $c_{Li} = c_{CF}^0$ for $\mathbf{x} \in \Omega_{CF}$, $c_{Li} = c_p^0$ for $\mathbf{x} \in \Omega_P$ and $c_{Li} = c_X = c_{ref}$ for $\mathbf{x} \in \Omega_{ED}$ at t = 0.

Time-incremental weak format of the full cell problem, current control

Upon employing the Backward–Euler rule, the galvanostatic problem for controlled current $I_{app}(t)$ is stated as follows: For known values of ${}^{n-1}c_{Li}$, ${}^{n}c_{X}$, we seek the spatial fields at time $t = t_n$: $\boldsymbol{u}, \varphi, \mu_{Li}, c_{Li}, \mu_X, c_X$, $\Phi_p \in \mathbb{U} \times \mathbb{F} \times \mathbb{M}_{Li} \times \mathbb{M}_X \times \mathbb{L}_2(\Omega_{ED}) \times \mathbb{R}$. We have used the notation $y(\boldsymbol{x}) = {}^n y(\boldsymbol{x}) = y(\boldsymbol{x}, t_n)$ and ${}^{n-1}y(\boldsymbol{x}) = y(\boldsymbol{x}, t_{n-1})$.

$$\int_{\Omega_{N}} \boldsymbol{\sigma} : \boldsymbol{\epsilon}[\delta \boldsymbol{u}] \, \mathrm{d}\, \Omega = 0 \quad \forall \delta \boldsymbol{u} \in \mathbb{U},$$
(19)

$$\int_{\Omega_{ED}} F[c_{Li} - c_X] \delta \varphi + \boldsymbol{d} \cdot [\nabla \delta \varphi] \, \mathrm{d}\,\Omega = -\int_{\Gamma_p^{\mathrm{int}}} \mathrm{d}_p \delta \varphi \, \mathrm{d}\,\Gamma + \int_{\Gamma_{CF}} \mathrm{d}_N \delta \varphi \, \mathrm{d}\,\Gamma \quad \forall \delta \varphi \in \mathbb{F},$$
(20)

$$\int_{\Omega} \frac{1}{\Delta t} c_{\mathrm{Li}} \delta \mu_{\mathrm{Li}} - \mathbf{j}_{\mathrm{Li}} \cdot [\nabla \delta \mu_{\mathrm{Li}}] \, \mathrm{d}\,\Omega - \int_{\Gamma_{\mathrm{int}}^{\mathrm{p}}} j_{\mathrm{p}} [\![\delta \mu_{\mathrm{Li}}]\!] \, \mathrm{d}\,\Gamma + \int_{\Gamma_{\mathrm{CF}}} j_{\mathrm{N}} [\![\delta \mu_{\mathrm{Li}}]\!] \, \mathrm{d}\,\Gamma$$
$$= \int_{\Omega} \frac{1}{\Delta t}^{n-1} c_{\mathrm{Li}} \delta \mu_{\mathrm{Li}} \, \mathrm{d}\,\Omega \quad \forall \delta \mu_{\mathrm{Li}} \in \mathbb{M}_{\mathrm{Li}},$$
(21)

$$\int_{\Omega_{ED}} \frac{1}{\Delta t} c_X \delta \mu_X - \mathbf{j}_X \cdot [\nabla \delta \mu_X] \, \mathrm{d}\,\Omega = \int_{\Omega_{ED}} \frac{1}{\Delta t} c_X \delta \mu_X \, \mathrm{d}\,\Omega \quad \forall \delta \mu_X \in \mathbb{M}_X,$$
(22)

$$\int_{\Omega_{CF} \cup \Omega_{ED} \cup \Omega_{P}} [\mu_{Li} - \mu_{Li}^{en}] \delta c_{Li} d\Omega = 0 \quad \forall \delta c_{Li} \in \mathbb{L}_{2}, \qquad (23)$$

$$\int_{\Omega_{ED}} [\mu_X - \mu_X^{en}] \delta c_X \, \mathrm{d}\, \Omega = 0 \quad \forall \delta c_X \in \mathbb{L}_2,$$
(24)

$$\left[I_{app} - F \int_{\Gamma_{p}^{int}} j_{p} \, \mathrm{d}\,\Gamma\right] \delta \Phi_{p} = 0 \quad \forall \delta \Phi_{p} \in \mathbb{R}.$$
(25)

The relevant solution (and test) spaces are defined as

$$\mathbb{U} = \left\{ \boldsymbol{u} \in \left[\mathbb{H}^{1}(\Omega_{\mathrm{N}}) \right]^{3} : \boldsymbol{u}^{+} - \boldsymbol{u}^{-} = \boldsymbol{0} \text{ on } \Gamma_{\mathrm{N}}^{\mathrm{ext},+}, \int_{\Omega_{\mathrm{N}}} \boldsymbol{u} \, \mathrm{d} \, \Omega = \boldsymbol{0} \right\},$$
(26)

$$\mathbb{F} = \left\{ \varphi \in \mathbb{H}^1(\Omega_{\text{ED}}) : \varphi^+ - \varphi^- = 0 \text{ on } \Gamma_{\text{N}}^+ \cup \Gamma_{\text{SEP}}^+ \right\},$$
(27)

$$\begin{split} \mathbb{M}_{\mathrm{Li}} &= \left\{ \mu \in \mathbb{L}_{2}(\Omega_{\mathrm{CF}} \cup \Omega_{\mathrm{ED}} \cup \Omega_{\mathrm{P}}) : \mu|_{\Omega_{\mathrm{CF}}} \in \mathbb{H}^{1}(\Omega_{\mathrm{CF}}), \mu|_{\Omega_{\mathrm{ED}}} \in \mathbb{H}^{1}(\Omega_{\mathrm{ED}}), \\ \mu|_{\Omega_{\mathrm{P}}} \in \mathbb{H}^{1}(\Omega_{\mathrm{P}}), \mu^{+} - \mu^{-} = 0 \text{ on } \Gamma_{\mathrm{N}}^{+} \cup \Gamma_{\mathrm{SEP}}^{+} \cup \Gamma_{\mathrm{P}}^{+} \right\}, \end{split}$$

$$(28)$$

$$\mathbb{M}_X = \left\{ \mu \in \mathbb{H}^1(\Omega_{\mathrm{ED}}) : \mu^+ - \mu^- = 0 \text{ on } \Gamma^+_{\mathrm{N}} \cup \Gamma^+_{\mathrm{SEP}} \right\}, \qquad (29)$$

where $\mathbb{L}_2(w)$ and $\mathbb{H}^1(w)$ denotes the space of square integrable functions, and the space of functions with square integrable derivatives of order 0 and 1, respectively.

Eq. (19) corresponds to the balance of linear momentum along with boundary conditions shown in Eqs. (1)–(3). Eq. (20) correspond to Gauss

law shown in Eq. (5) combined with boundary conditions in Eqs. (8)–(10). The weak representation of Li-mass balance in Eq. (21) originates from its strong form counterparts in Eqs. (4) and (12), along with the relevant boundary conditions in Eqs. (6), (8)–(10) and (13)–(16). Similarly, the weak representation of the anion mass balance in Eq. (22), is related to its strong counterpart in Eq. (4) with boundary conditions in Eqs. (6) and (11). Finally, Eqs. (23) and (24) enforce the chemical potential, μ_{av} to follow the constitutive relation $\mu_{\alpha}^{\text{en}} = \mu_{\alpha}^{\text{en}}(c_{\alpha})$ and Eq. (25) forces the electric current over Γ_{P}^{int} to follow the prescribed current in I_{app} . We recall that the negative electrode potential is set to (reference) $\Phi_{N} = 0$. In the following sections, we introduce constitutive relations for $\boldsymbol{\sigma}, \mu_{\alpha}^{\text{en}}, j_{\alpha}, j_{N}, j_{P}, \boldsymbol{d}, d_{N}$ and d_{P} .

Constitutive relations for the carbon fibre domain in the negative electrode

Lithiation of carbon fibres are accompanied by a change in moduli and an anisotropic expansion characterised by $\alpha_{\rm CF}$, containing longitudinal and transverse expansion coefficients $\alpha_{\rm L}$ and $\alpha_{\rm T}$, respectively^{6,15}. The Helmholtz free energy for the carbon fibres is expressed as

$$\psi_{\rm CF}(\epsilon, c_{\rm Li}) = \frac{1}{2} \left[\epsilon - \alpha_{\rm CF} \frac{c_{\rm Li} - c_{\rm CF}^0}{\tilde{c}_{\rm CF}} \right] : \mathbf{E}_{\rm CF}(c_{\rm Li}) : \left[\epsilon - \alpha_{\rm CF} \frac{c_{\rm Li} - c_{\rm CF}^0}{\tilde{c}_{\rm CF}} \right] + \bar{\psi}(c_{\rm Li}, \tilde{c}_{\rm CF} - c_{\rm Li}),$$
(30)

where $\mathbf{E}_{CF}(c_{Li})$ represents the fourth order (anisotropic) stiffness tensor of the carbon fibres described in Larsson et al.¹⁶. The relevant material parameters related to $\mathbf{E}_{CF}(c_{Li})$ are the following moduli and Poisson ratios $E_L(c_{Li}), E_T(c_{Li}), v_{LT}, v_{TT}, G_{LT}$ and G_{TT} . Furthermore, $\boldsymbol{\epsilon}(\boldsymbol{u})$ is the (small) strain tensor. The maximum (reversible) concentration can be defined as the sum of inserted lithium concentration and the concentration of vacant sites, $\tilde{c}_{CF} = c_v + c_{Li}$, where c_v is the concentration of vacant sites for possible lithium insertion. In this work, we consider conditioned carbon fibres, assuming the maximum concentration is constant. Consequently, no Lidegradation mechanisms in the electrode such as lithium trapping, dendrite growth, or SEI formation are considered. The chemical contribution to the free energy is expressed as

$$\bar{\psi}(c_{\rm Li}, c_{\rm v}) = c_{\rm Li} \mu_{\rm Li,CF}^0 + c_{\rm v} \mu_{\rm v,CF}^0 + c_{\rm Li} RT \ln(c_{\rm Li}) + c_{\rm v} RT \ln(c_{\rm v}) + \Lambda(c_{\rm Li}, c_{\rm v}),$$
(31)

where *R* is the ideal gas constant and *T* is the temperature. $\Lambda(c_{\text{Li}}, c_{\text{v}})$ represents the deviation from ideal conditions caused by interaction of inserted lithium and the vacant sites. We adopt a formulation similar to Landstorfer et al.¹⁷, with the solubility parameter k_{CF} so that

$$\frac{\mathrm{d}}{\mathrm{d}\,c_{\mathrm{Li}}}\Lambda(c_{\mathrm{Li}},\widetilde{c}_{\mathrm{CF}}-c_{\mathrm{Li}}) = RTk_{\mathrm{CF}}\left(1-\frac{c_{\mathrm{Li}}}{\widetilde{c}_{\mathrm{CF}}}\right). \tag{32}$$

We can now express the following constitutive relations

$$\boldsymbol{\sigma}(\boldsymbol{\epsilon}, c_{\mathrm{Li}}) = \frac{\partial \psi_{\mathrm{CF}}}{\partial \boldsymbol{\epsilon}} = \mathbf{E}_{\mathrm{CF}}(c_{\mathrm{Li}}) : \left[\boldsymbol{\epsilon} - \boldsymbol{\alpha}_{\mathrm{CF}} \frac{c_{\mathrm{Li}}}{\tilde{c}_{\mathrm{CF}}}\right], \quad (33)$$

$$\mu_{\mathrm{Li}}^{\mathrm{en}}(\boldsymbol{\epsilon}, c_{\mathrm{Li}}) = \frac{\partial \psi_{\mathrm{CF}}}{\partial c_{\mathrm{Li}}} = -\frac{1}{\widetilde{c}_{\mathrm{CF}}} \boldsymbol{\alpha}_{\mathrm{CF}} : \boldsymbol{\sigma} + \mu_{\mathrm{CF}}^{0} + RT \left[\ln \left(\frac{c_{\mathrm{Li}}}{\widetilde{c}_{\mathrm{CF}} - c_{\mathrm{Li}}} \right) + k_{\mathrm{CF}} \left(1 - \frac{c_{\mathrm{Li}}}{\widetilde{c}_{\mathrm{CF}}} \right) \right],$$
(34)

where the carbon fibre reference chemical potential for the carbon fibres is obtained as $\mu_{CF}^0 = \mu_{Li,CF}^0 - \mu_{\nu,CF}^0$, $\mu_{Li,CF}^0$ and $\mu_{\nu,CF}^0$ are the reference chemical potentials for lithium and vacancies, respectively. The contribution from the concentration dependent stiffness is ignored. The mass flux of neutral lithium within the carbon fibres is governed by the gradient of chemical potential scaled by an isotropic mobility

$$\boldsymbol{j}_{\mathrm{Li}} = -M_{\mathrm{Li}}(\boldsymbol{c}_{\mathrm{Li}})\nabla\boldsymbol{\mu}_{\mathrm{Li}}.$$
(35)

The mobility along the fibre (1-dir) is not equal to the mobility in the transversely isotropic plane (2-3-dir). Here, we assign the isotropic mobility equal to the mobility in the 2-3-dir.

Constitutive relations for the structural battery electrolyte domain

The porous, bi-phasic structural battery electrolyte matrix enables ion transport as well as mechanical load transfer. Recent studies carried out by Duan et al.¹⁸ show that the pores are homogeneously distributed and that the SBE is isotropic. We model the SBE using linear isotropic elasticity and introduce the Helmholtz free energy for the SBE domain as

$$\psi_{\text{SBE}}(\boldsymbol{\epsilon}, c_{\text{Li}}, c_X, \nabla \varphi) = \frac{1}{2} \boldsymbol{\epsilon} : \mathbf{E}_{\text{SBE}} : \boldsymbol{\epsilon} + c_{\text{Li}} \left[\mu_{\text{Li}}^0 + RT \ln\left(\frac{c_{\text{Li}}}{c_{\text{ref}}}\right) - RT \right] \\ + c_X \left[\mu_X^0 + RT \ln\left(\frac{c_X}{c_{\text{ref}}}\right) - RT \right] - \frac{1}{2} \boldsymbol{\epsilon} (\nabla \varphi)^2,$$
(36)

where \mathbf{E}_{SBE} is the homogeneous isotropic fourth-order stiffness tensor with related shear and bulk moduli *G* and *K*. c_{ref} is the salt concentration in the liquid electrolyte phase, ε is the electric permittivity, $\nabla \varphi$ the electric field and μ_{α}^{α} the reference chemical potential of lithium ions ($\alpha = \text{Li}$) and the anions ($\alpha = X$). The following (decoupled) constitutive relations can be derived

$$\boldsymbol{\sigma}(\boldsymbol{\epsilon}) = \frac{\partial \psi_{\text{SBE}}}{\partial \boldsymbol{\epsilon}} = \mathbf{E}_{\text{SBE}} : \boldsymbol{\epsilon} = 2G \operatorname{dev}(\boldsymbol{\epsilon}) + \operatorname{K} \operatorname{tr}(\boldsymbol{\epsilon}) \boldsymbol{I}, \quad (37)$$

$$\mu_{\rm Li}^{\rm en}(c_{\rm Li}) = \frac{\partial \psi_{\rm SBE}}{\partial c_{\rm Li}} = \mu_{\rm Li}^0 + RT \ln\left(\frac{c_{\rm Li}}{c_{\rm ref}}\right),\tag{38}$$

$$\mu_X^{\rm en}(c_X) = \frac{\partial \psi_{\rm SBE}}{\partial c_X} = \mu_X^0 + RT \ln\left(\frac{c_X}{c_{\rm ref}}\right),\tag{39}$$

$$\boldsymbol{d}(\boldsymbol{\varphi}) = \frac{\partial \boldsymbol{\psi}}{\partial \nabla \boldsymbol{\varphi}} = -\varepsilon \nabla \boldsymbol{\varphi}.$$
(40)

The mass flux of Li ions and accompanying anion are expressed as

$$\mathbf{j}_{\mathrm{Li}}(\mu_{\mathrm{Li}}, \nabla \varphi) = -\eta_{\mathrm{Li}} c_{\mathrm{Li}} [\nabla \mu_{\mathrm{Li}} + F \nabla \varphi], \tag{41}$$

$$\boldsymbol{j}_{\boldsymbol{X}}(\boldsymbol{\mu}_{\boldsymbol{X}}, \nabla \boldsymbol{\varphi}) = -\eta_{\boldsymbol{X}} \boldsymbol{c}_{\boldsymbol{X}} [\nabla \boldsymbol{\mu}_{\boldsymbol{X}} - F \nabla \boldsymbol{\varphi}], \tag{42}$$

where η_{Li} and η_X are the isotropic mobility coefficients of lithium ions and anions, respectively. We note that both chemical and electric potential gradients contribute to ionic transport, i.e. diffusion and migration. The electric field affects the ionic transport of the cation and anion in opposite directions due to the different charge of the ions. The mobilities of Eqs. (41) and (42) follow from the assumption of constant diffusion coefficients in the electrolyte.

Constitutive relations for the separator domain

Similar to the SBE, the same equations can be applied over the separator domain. The separator consists of a mixture of SBE and isotropic glass fibre separator. The volume fraction of glass fibre separator is estimated as 34% using available data from Asp et al.². The effective mobilities and the elasticity tensor are obtained using volume averaging, where we assume that the glass fibres block ionic transport. The effective mobilities thus become $\bar{\eta}_{\rm Li} = (1 - V_{\rm GF})\eta_{\rm Li}$, $\bar{\eta}_X = (1 - V_{\rm GF})\eta_X$ and $\mathbf{E}_{\rm SEP} = (1 - V_{\rm GF})\mathbf{E}_{\rm SBE} + V_{\rm GF}\mathbf{E}_{\rm GF}$, where $V_{\rm GF}$ is the volume fraction of glass fibres in the separator. The

Helmholtz free energy applicable to Ω_{SEP} is expressed as

$$\psi_{\text{SEP}}(\boldsymbol{\epsilon}, c_{\text{Li}}, c_X, \nabla \varphi) = \frac{1}{2} \boldsymbol{\epsilon} : \mathbf{E}_{\text{SEP}} : \boldsymbol{\epsilon} + c_{\text{Li}} \left[\mu_{\text{Li}}^0 + RT \ln\left(\frac{c_{\text{Li}}}{c_{\text{ref}}}\right) - RT \right] \\ + c_X \left[\mu_X^0 + RT \ln\left(\frac{c_X}{c_{\text{ref}}}\right) - RT \right] - \frac{1}{2} \boldsymbol{\epsilon} (\nabla \varphi)^2,$$
(43)

The constitutive relations are expressed as

$$\sigma(\boldsymbol{\epsilon}) = \frac{\partial \psi_{\text{SEP}}}{\partial \boldsymbol{\epsilon}} = \mathbf{E}_{\text{SEP}} : \boldsymbol{\epsilon} = 2\bar{\text{G}}\,\text{dev}\,(\boldsymbol{\epsilon}) + \bar{K}\,\text{tr}\,(\boldsymbol{\epsilon})\boldsymbol{I},\tag{44}$$

$$\mu_{\text{Li}}^{\text{en}}(c_{\text{Li}}) = \frac{\partial \psi_{\text{SEP}}}{\partial c_{\text{Li}}} = \mu_{\text{Li}}^0 + RT \ln\left(\frac{c_{\text{Li}}}{c_{\text{ref}}}\right),\tag{45}$$

$$\mu_X^{\rm en}(c_X) = \frac{\partial \psi_{\rm SEP}}{\partial c_X} = \mu_X^0 + RT \ln\left(\frac{c_X}{c_{\rm ref}}\right),\tag{46}$$

$$\boldsymbol{d}(\boldsymbol{\varphi}) = \frac{\partial \psi_{\text{SEP}}}{\partial \nabla \boldsymbol{\varphi}} = -\varepsilon \nabla \boldsymbol{\varphi}, \qquad (47)$$

$$\mathbf{j}_{\mathrm{Li}}(\mu_{\mathrm{Li}}, \nabla \varphi) = -\bar{\eta}_{\mathrm{Li}} c_{\mathrm{Li}} [\nabla \mu_{\mathrm{Li}} + F \nabla \varphi], \qquad (48)$$

$$\boldsymbol{j}_{\boldsymbol{X}}(\boldsymbol{\mu}_{\boldsymbol{X}}, \nabla \boldsymbol{\varphi}) = -\bar{\boldsymbol{\eta}}_{\boldsymbol{X}} \boldsymbol{c}_{\boldsymbol{X}} [\nabla \boldsymbol{\mu}_{\boldsymbol{X}} - F \nabla \boldsymbol{\varphi}]. \tag{49}$$

Constitutive relations for the positive electrode domain

We adopt the same formulation as introduced in the section "Introduction" for the positive electrode. It is important to note that data pertaining to the homogeneous expansion of the positive electrode is unavailable. This choice effectively disables the coupling of chemo-mechanical fields in the positive electrode, and we consider the positive electrode as stress-free upon lithium insertion. It is acknowledged that $\alpha_{\rm P}$ can be obtained through measurements or by employing computational homogenisation^{16,19}. The elastic properties of a positive electrode slurry were measured by Gupta et al.²⁰. They reported a tensile modulus of 0.90 GPa for the NMC based positive electrode continuum. We base the elastic properties of the homogenised positive electrode and solubility parameter of the positive electrode are denoted as $\tilde{c}_{\rm P}$ and $k_{\rm P}$, respectively. The Helmholtz free energy of the positive electrode is expressed as

$$\begin{split} \psi_{\mathrm{P}}(\boldsymbol{\epsilon}, c_{\mathrm{Li}}) &= \frac{1}{2} \left[\boldsymbol{\epsilon} - \boldsymbol{\alpha}_{\mathrm{P}} \frac{c_{\mathrm{Li}} - c_{\mathrm{P}}^{0}}{\tilde{c}_{\mathrm{P}}} \right] : \mathbf{E}_{\mathrm{P}}(c_{\mathrm{Li}}) \\ &: \left[\boldsymbol{\epsilon} - \boldsymbol{\alpha}_{\mathrm{P}} \frac{c_{\mathrm{Li}} - c_{\mathrm{P}}^{0}}{\tilde{c}_{\mathrm{P}}} \right] + \bar{\psi}(c_{\mathrm{Li}}, \tilde{c}_{\mathrm{P}} - c_{\mathrm{Li}}), \end{split}$$
(50)

where

$$\bar{\psi}(c_{\rm Li}, c_{\rm v}) = c_{\rm Li} \mu_{\rm Li,P}^0 + c_{\rm v} \mu_{\rm v,P}^0 + c_{\rm Li} RT \ln(c_{\rm Li}) + c_{\rm v} RT \ln(c_{\rm v}) + \Lambda(c_{\rm Li}, c_{\rm v}).$$
(51)

and

$$\frac{\mathrm{d}}{\mathrm{d}\,c_{\mathrm{Li}}}\Lambda(c_{\mathrm{Li}},\widetilde{c}_{\mathrm{P}}-c_{\mathrm{Li}}) = RTk_{\mathrm{P}}\left(1-\frac{c_{\mathrm{Li}}}{\widetilde{c}_{\mathrm{P}}}\right). \tag{52}$$

The constitutive relations in the positive electrode can be expressed as

$$\boldsymbol{\sigma}(\boldsymbol{\epsilon}, c_{\mathrm{Li}}) = \frac{\partial \psi_{\mathrm{P}}}{\partial \boldsymbol{\epsilon}} = \mathbf{E}_{\mathrm{P}}(c_{\mathrm{Li}}) : \left[\boldsymbol{\epsilon} - \boldsymbol{\alpha}_{\mathrm{P}} \frac{c_{\mathrm{Li}}}{\tilde{c}_{\mathrm{P}}}\right], \tag{53}$$



Fig. 6 | Fluxes over generic interface. Illustration of a generic electrode/electrolyte

$$\mu_{\mathrm{Li}}^{\mathrm{en}}(\boldsymbol{\epsilon}, c_{\mathrm{Li}}) = \frac{\partial \psi_{\mathrm{P}}}{\partial c_{\mathrm{Li}}} = \mu_{\mathrm{P}}^{0} + RT \left[\ln \left(\frac{c_{\mathrm{Li}}}{\widetilde{c}_{\mathrm{P}} - c_{\mathrm{Li}}} \right) + k_{\mathrm{P}} \left(1 - \frac{c_{\mathrm{Li}}}{\widetilde{c}_{\mathrm{P}}} \right) \right].$$
(54)

The mass flux in the positive electrode is characterised by the effective diffusion coefficient $D_{\text{Li},\text{P}}$, representing the homogenised underlying transport mechanisms in the positive electrode slurry. The mobility of neutral lithium in the positive electrode is derived assuming a constant diffusion coefficient such that the mass flux becomes

$$\boldsymbol{j}_{\mathrm{Li}} = \boldsymbol{D}_{\mathrm{Li},\mathrm{P}} \left[\frac{\partial \boldsymbol{\mu}_{\mathrm{Li}}}{\partial \boldsymbol{c}} \right]^{-1} \nabla \boldsymbol{\mu}_{\mathrm{Li}}, \tag{55}$$

where $\frac{\partial \mu_{\text{Li}}}{\partial c}$ is computed from Eq. (54).

interface.

Constitutive relations for electrode-electrolyte interfaces

Figure 6 shows a generic electrode/electrolyte interface where Γ describes the transition from the electrode (- side) to the electrolyte (+ side). The interface mass flux of lithium ions is denoted $j_n = \mathbf{j}_{Li} \cdot \mathbf{n}_{\Gamma}$, where \mathbf{n}_{Γ} is the unit normal pointing from (-) to (+). We consider the case where no lithium ions accumulates on Γ , whereby $j_n^- = j_n^+ = j_n$. Furthermore, we do not allow accumulation of free charge on the boundary, whereby the current $i_n^- = i_n^+ = i_n$ also becomes continuous. Since there is no flux of anions across Γ , we conclude that

$$i_{\rm n}^+ = F j_{\rm n}^+. \tag{56}$$

Assuming no net free charge on the interface, we also obtain continuity in the electric flux densities $d_n^- = d_n^+ = d_n$. Finally, for an isotropic ideal conductor, the electric flux density can be expressed as

$$\boldsymbol{d}_{n}^{-} = -\varepsilon_{c} [\nabla \boldsymbol{\varphi}]^{-} \cdot \boldsymbol{n}_{\Gamma} = -\frac{\varepsilon_{c}}{\kappa_{c}} \kappa_{c} [\nabla \boldsymbol{\varphi}]^{-} \cdot \boldsymbol{n}_{\Gamma} = \frac{\varepsilon_{c}}{\kappa_{c}} i_{n}^{-}, \qquad (57)$$

at the electrode side, where ε_c and κ_c are the electric permittivity and conductivity of the homogeneous electrode. Using Eqs. (56) and (57) for the continuous fluxes, we arrive at

$$i_{\rm n} = F j_{\rm n}, \tag{58}$$

$$d_{\rm n} = \frac{\varepsilon_{\rm c}}{\kappa_{\rm c}} F j_{\rm n}.$$
 (59)

The interface mass flux over the positive and negative electrode/electrolyte interfaces is described by the Butler–Volmer relation, where the mass flux is continuous over the interface while the electric and chemical potential fields are discontinuous. The interface relation over positive electrode and separator interface is expressed as

$$j_{\rm P}(\eta_{\rm P}) = -\frac{i_{0,\rm P}(c_{\rm Li})}{F} \left[\exp\left(\frac{\eta_{\rm P}}{2RT}\right) - \exp\left(\frac{-\eta_{\rm P}}{2RT}\right) \right],\tag{60}$$

where the overpotential

$$\eta_{\rm P} = \llbracket \mu_{\rm Li} \rrbracket + F \llbracket \varphi \rrbracket = \llbracket \mu_{\rm Li} \rrbracket + F \llbracket \varphi^+ - \Phi_{\rm P} \rrbracket, \tag{61}$$

and the interface electric flux density is expressed as

$$d_{\rm P}(\eta_{\rm P}) = \frac{\varepsilon_{\rm P}}{\kappa_{\rm P}} F j_{\rm P}(\eta_{\rm P}). \tag{62}$$

Here, $i_{0,P}(c_{Li})$ is the exchange current density parametrised in the electrode concentration c_{Li} , pertinent to the positive electrode/separator interface expressed as a third degree polynomial with coefficients q_i so that $i_{0,P}(c_{Li}) = q_0 + q_1c_{Li} + q_2c_{Li}^2 + q_3c_{Li}^3$. Furthermore, assuming a nearconstant concentration in the electrolyte. Φ_P is the electric potential, ε_P is the electric permittivity and κ_P is the electric conductivity of the positive electrode. Similarly, the interface mass flux and electric charge flux density over the carbon fibres in the negative electrode and SBE interfaces are expressed as

$$\dot{\eta}_{\rm N}(\eta_{\rm N}) = -\frac{\dot{i}_{0,\rm N}(c_{\rm Li})}{F} \Big[\exp\left(\frac{\eta_{\rm N}}{2RT}\right) - \exp\left(\frac{-\eta_{\rm N}}{2RT}\right) \Big],\tag{63}$$

$$d_{\rm N}(\eta_{\rm N}) = \frac{\varepsilon_{\rm N}}{\kappa_{\rm N}} F j_{\rm N}(\eta_{\rm N}), \tag{64}$$

with the overpotential

$$\eta_{\rm N} = [\![\mu_{\rm Li}]\!] + F[\![\phi]\!] = [\![\mu_{\rm Li}]\!] + F[\Phi_{\rm N} - \phi^-].$$
(65)

 $i_{0,N}(c_{Li})$ is the exchange current density of the SBE/carbon fibre interface parametrised in local carbon fibre Li-concentration, Φ_N is the electric potential in the negative electrode, ε_N is the electric permittivity and κ_N is the electric conductivity of the negative electrode.

Experimental overview

This section addresses the determination of material parameters aimed at minimising the difference between the experimental charge/discharge curves and simulated response voltage profiles for the same charge rates. Two unique cells are considered in the calibration and validation process. The first cell is manufactured in-house following the process described by Siraj et al.¹ and contains approximately 12,000 T800 carbon fibres in the negative electrode. Furthermore, the cell extends 33 mm the fibre direction and is cycled at C/20 and C/10, including a 30 min rest time. The second cell was manufactured at a different point in time by Siraj et al.¹ and contains ~24,000 fibres with a length of 50 mm in the fibre direction. The charge–discharge data from the second cell, cycled at C/2, is used to validate the calibrated parameters. The applied current related to each cell and C-rate, is normalised with the calculated weight of carbon fibres in the cell where $\rho_{\rm CF} = 1750$ kg/m³ is the density of the carbon fibres. These currents are applied to the model and given as a function of time through the variable $I_{\rm app}$ in Eq. (25).

Identification of carbon fibre electrode properties

Kjell et al.²¹ conducted measurements of the diffusion coefficient and exchange current density of a single IMS65 carbon fibre in relation to the state of charge. To reduce the number of calibration parameters, the

diffusion coefficient and exchange current density data is utilised to determine expressions for the equivalent mobility and exchange current density for the negative electrode. The measurements on the carbon fibre was performed in liquid electrolyte, which implies stress free expansion of the carbon fibre as lithium inserts.

$$\mathbf{j}_{\mathrm{Li}} = -M_{\mathrm{Li,CF}}(c_{\mathrm{Li}})\nabla\mu_{\mathrm{Li}} = -M_{\mathrm{Li,CF}}(c_{\mathrm{Li}})\frac{\partial\mu^{\mathrm{en}}}{\partial c_{\mathrm{Li}}}\Big|_{\boldsymbol{\sigma}=\mathbf{0}}\nabla c_{\mathrm{Li}} = -D_{\mathrm{Li,CF}}(c_{\mathrm{Li}})\nabla c_{\mathrm{Li}}.$$
(66)

By identifying $M_{\text{Li,CF}}(c_{\text{Li}}) = \left(\frac{\partial \mu^{e_{\text{I}}}}{\partial c_{\text{Li}}}\right)^{-1} D_{\text{Li,CF}}(c_{\text{Li}})$, where $D_{\text{Li,CF}}^{\text{M}}$ contains discrete data and $\left(\frac{\partial \mu^{e_{\text{I}}}}{\partial c_{\text{Li}}}\right)^{-1}$ is obtained from Eq. (34), we define $D_{\text{Li,CF}}^{*}(c_{\text{Li}})$ as a second-degree polynomial determined by

$$D_{\text{Li,CF}}^{*}(c_{\text{Li}}) = \arg\min_{P^{2}([0, \tilde{c}_{\text{CF}}])} ||D_{\text{Li,CF}}(c_{\text{Li}}) - D_{\text{Li,CF}}^{M}||.$$
(67)

Thereby, the mass flux in the carbon fibres is expressed as

$$\mathbf{j}_{\mathrm{Li}} = -\left[\frac{\partial \mu^{\mathrm{en}}(\boldsymbol{\epsilon}, c_{\mathrm{Li}})}{\partial c_{\mathrm{Li}}}\right]_{\boldsymbol{\sigma}=\mathbf{0}}^{-1} D^*_{\mathrm{Li},\mathrm{CF}}(c_{\mathrm{Li}}) \nabla \mu_{\mathrm{Li}}.$$
(68)

Similarly, the exchange current density data is approximated by a thirddegree polynomial determined by minimising the function

$$i_{0,N}^{*}(c_{\text{Li}}) = \arg \min_{p^{3}([0,\widetilde{c}_{\text{CF}}])} \left\| i_{0,N}(c_{\text{Li}}) - i_{0,N}^{M} \right\|.$$
(69)

The resulting polynomial fits are shown in Fig. 7 where the solid curves represent the discrete data points and the dashed curves are the continuous functions obtained using Eqs. (67) and (69).

Furthermore, the reference carbon fibre chemical potential μ_{CF}^0 is obtained from measurements by Carlstedt et al.⁶, where the carbon fibre negative electrode is analysed in a half-cell configuration vs. lithium metal. The equilibrium potential of the half-cell was measured across various concentrations. We utilise the experimental measurements to uniquely determine the magnitude of the reference chemical potential μ_{CF}^0 . In a half-cell configuration, the carbon fibres serve as the positive electrode denoted \bullet_{CF} , while lithium metal acts as the negative electrode denoted \bullet_{ref} . Furthermore, quantities related to the electrolyte are denoted without subscript. At net zero current, the following conditions hold:

$$j_{\rm ref} = j_{\rm CF} = 0, \eta_{\rm ref} = \eta_{\rm CF} = 0$$
 (70)

Here, $\eta_{CF} = F[\varphi - \Phi_{CF}] + \mu - \mu_{CF}$ and $\eta_{ref} = F[\Phi_{ref} - \varphi] + \mu_{ref} - \mu$ are the overpotentials at carbon fibre / electrolyte and lithium metal/electrolyte interfaces, respectively. After relaxation, at zero current, a stationary electric potential is obtained. Additionally, the concentration of excess lithium in the electrolyte inserts into the carbon fibres, leading to $c_{Li} = c_{ref}$ in the electrolyte. Moreover, the reference chemical potential in the electrolyte is assumed to be zero. Setting the electric potential of the negative electrode as the reference, the overpotentials at each interface can be expressed as

$$\eta_{\rm CF} = F[\varphi - \Phi_{\rm CF}] - \mu_{\rm CF},\tag{71}$$

$$\eta_{\rm ref} = F[0 - \varphi] + \mu_{\rm ref}.$$
(72)

The chemical potential of lithium metal is not significantly affected by concentration variations, i.e., the activity of the solid phase is one, $\mu_{\rm ref} = \mu_{\rm ref}^{0}$ ²². From Eq. (72), $\eta_{\rm ref} = 0$ gives the electrolyte potential

$$\varphi = \frac{1}{F} \mu_{\rm ref}^0. \tag{73}$$

Fig. 7 | Diffusion and exchange current

density fits. a Diffusion coefficient discrete data from Kjell et al.²¹ and the polynomial fit as a function of normalised concentration. **b** Exchange current density discrete data from Kjell et al.²¹ and the related polynomial fit as a function of normalised concentration



We adopt Eq. (34) as the model for the chemical potential in the carbon fibre. Assuming a a stress free fibre together with Eq. (73) together with $\eta^{\text{CF}} = 0$ in Eq. (72) gives

$$\mu_{\rm CF}^0 = \mu_{\rm ref}^0 - F \Phi_{\rm CF} - RT \left[\ln \left(\frac{c_{\rm Li}}{\tilde{c}_{\rm CF} - c_{\rm Li}} \right) + k_{\rm CF} \left(1 - \frac{c_{\rm Li}}{\tilde{c}_{\rm CF}} \right) \right], \quad (74)$$

where $c_{\rm Li}$ is the Li-concentration in the fibre. The reference chemical potential of lithium metal phase approximated as $\mu^0_{\rm ref} = 0$, see for instance Mayur et al. or work carried out by Lai et al.^{23,24}. At $c_{\rm Li}/\tilde{c}_{\rm CF} = 0.5$ the measured equilibrium potential was $\Phi^{0.5}_{\rm CF} = 0.2$ V⁶.

$$\Phi_{\rm CF}^{0.5}F = \mu_{\rm ref}^0 - \mu_{\rm CF}^0 - \frac{1}{2}RTk_{\rm CF},\tag{75}$$

thereby, the reference chemical potential of the fibre can be expressed as

$$\mu_{\rm CF}^0 = -\Phi_{\rm CF}^{0.5} F - \frac{RTk_{\rm CF}}{2}.$$
(76)

Hence, $\mu_{\rm CF}^0$ is determined uniquely from the coefficient $k_{\rm CF}$.

SBE conductivity

The ionic conductivity of the SBE denoted κ_{SBE} , can be related to the mobility coefficients of the cations and anions under the assumption that no concentration gradients are present, and $c_{\text{Li}} \approx c_X \approx c_{\text{ref}}$ is expressed as follows

$$\boldsymbol{i} = \sum_{\alpha} z_{\alpha} F \boldsymbol{j}_{\alpha} = -\sum_{\alpha} z_{\alpha} F(\eta_{\alpha} c_{\alpha} z_{\alpha}) F \nabla \varphi = -\kappa_{\text{SBE}} \nabla \varphi, \qquad (77)$$

by identification, the ionic conductivity in the SBE can be expressed as

$$\kappa_{\rm SBE} = F^2 \sum_{\alpha} z_{\alpha}^2 \eta_{\alpha} c_{\alpha} = F^2 (\eta_{\rm Li} c_{\rm Li} + \eta_X c_X) = F^2 c_{\rm ref} (\eta_{\rm Li} + \eta_X), \quad (78)$$

where z_{α} is the charge number of the ion. Additionally, the transference number t^+ relates the mobility coefficients of the cation and anion to each other, we express

$$t_{+} = \frac{\eta_{\rm Li}}{\eta_{\rm Li} + \eta_{\rm X}},\tag{79}$$

$$\kappa_{\rm SBE} = \frac{1}{t_+} F^2 c_{\rm ref} \eta_{\rm Li}.$$
 (80)

Table 4 | Set of parameters for calibration

Index	Parameter	Description
θ_1	μ_{P}^{0}	Reference chemical potential in the positive electrode
θ2	k _P	Solubility parameter in the positive electrode
θ_3	k _{CF}	Solubility parameter in the carbon fibres
θ_4	<i>q</i> ₀	Parameter related to exchange current density of the positive electrode
θ_5	<i>q</i> ₁	Parameter related to exchange current density of the positive electrode
θ_6	q ₂	Parameter related to exchange current density of the positive electrode
θ_7	q ₃	Parameter related to exchange current density of the positive electrode
θ_8	€ C _P	Maximum concentration in the positive electrode
θ_9	\widetilde{c}_{CF}	Maximum concentration in the carbon fibres
θ_{10}	D _{Li,P}	Effective diffusion coefficient in the positive electrode
θ_{11}	КР	Effective electric conductivity in the positive electrode
θ ₁₂	KSBE	lonic conductivity in the electrolyte
θ_{13}	t_+	Transference number

In this way, both the mobility of lithium and the related anion can be uniquely determined. Cattaruzza et al.¹³ conducted measurements regarding the ionic conductivity, κ_{SBE} , and transference number, t^+ , for various salt concentrations and electrolyte volume fractions. They report a variation in conductivity, ranging from 0.037 mS/cm at 40 wt% electrolyte content to 0.29 mS/cm at 50 wt% electrolyte content. Meanwhile, the transference number exhibits a slight increase, moving from 0.34 to 0.43 with an increase in electrolyte content. The reference salt concentration of the SBE liquid phase remained constant at 1000 mol/m³ throughout these measurements. For the cells manufactured by Siraj et al.¹, the SBE contained 50 wt% liquid electrolyte.

Parameter identification

To identify the remaining material parameters we utilise the experimental charge-rest-discharge profiles from the first cell described in section "Introduction". The optimal material parameters, θ^* , are determined as the following minimiser

$$\boldsymbol{\theta}^{*} = \arg\min_{\boldsymbol{\theta}} \left[N_{C/10} \sum_{n=1}^{N_{C/20}} \left(\Phi_{\exp,C/20}(t_{n}) - \Phi^{+}(\boldsymbol{\theta}, t_{n}) \right)^{2} + N_{C/20} \sum_{n=1}^{N_{C/10}} \left(\Phi_{\exp,C/10}(t_{n}) - \Phi^{+}(\boldsymbol{\theta}, t_{n}) \right)^{2} \right]$$
(81)

Here, $\Phi_{\exp,C/20}(t)$ and $\Phi_{\exp,C/10}(t)$ is the experimental cell potentials over time, containing $N_{C/20}$ and $N_{C/10}$ data points, respectively. The material parameters, θ , considered in the calibration are collected in Table 4.

Although the ionic conductivity and transference number in the SBE are measured parameters, it is evident that they are sensitive to measurement errors based on the spread of the parameters with respect to the weight percent of solid phase¹³. Therefore, we include κ_{SBE} and t^+ in the parameter set, where variations within the measured range is allowed. Once θ^* is obtained, we assess the sensitivity of the simulated cell potential to perturbations in each material parameter. The purpose of the sensitivity analysis is to quantify how the model parameters affect the obtained, simulated cell potential over time. The sensitivity of parameter *i* is computed as

$$\Delta_i \Phi^+(\boldsymbol{\theta}^*, t) = \Phi^+(\boldsymbol{\theta}^* + h_i \boldsymbol{e}_i, t) - \Phi^+(\boldsymbol{\theta}^*, t)$$
(82)

where h_i is the magnitude of the perturbation and $(e_i)_i = \delta_{ii}$.

Data availability

Data will be made available upon request.

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Author contributions

C.L. wrote the manuscript, performed computations, conceptualised the methods, prepared figures, performed experiments. F.L. wrote the manuscript, supervision, conceptualised the methods. J.X. performed experiments, data processing. K.R. conceptualised methods. L.A. wrote the manuscript, supervision, conceptualised methods. All authors reviewed the manuscript.

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Competing interests

The authors declare no competing interests.

Additional information

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