



A physics-based model explaining stress-induced degradation in composite cathode for all-solid-state batteries[☆]

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ARTICLE INFO

Keywords:

Solid-state battery
Degradation
Model-based simulation

ABSTRACT

All-solid-state batteries (ASSBs) offer improved safety and energy density compared to conventional liquid-electrolyte systems, yet their widespread adoption is hindered by complex electro-chemo-mechanical degradation mechanisms. In this work, we introduce a stochastic continuum framework to capture microstructural heterogeneity and its influence on interfacial degradation in ASSBs. The approach combines physics-based modeling of electro-chemo-mechanical coupling, including a traction-separation law for interface decohesion, with non-Gaussian, spatially correlated random fields representing the variability of solid electrolyte properties. Random fields are constructed via a Gaussian translation method with Gamma marginals and a copula-based correlation structure, enabling realistic representation of particle-scale property fluctuations while maintaining computational efficiency. Numerical simulations on a representative half-cell of NMC811 cathode particles embedded in LPSCl electrolyte demonstrate the emergence of localized stress concentrations and interface damage under cyclic charging. The model quantitatively captures the evolution of specific differential capacity and depth of discharge, revealing how heterogeneous electrolyte properties amplify interfacial failure and drive catastrophic degradation. This framework provides a versatile tool for predictive design and reliability assessment of ASSBs accounting for microstructural variability.

1. Introduction

All-solid-state batteries (ASSBs) have emerged as a next-generation energy storage technology owing to their potential for improved safety, higher energy density, and wider operating temperature ranges compared to conventional liquid-electrolyte systems [1–3]. Despite these advantages, practical deployment of ASSBs remains limited by a variety of degradation mechanisms that critically affect performance and cycle life. Among the most prominent challenges are interfacial instability between electrode and solid electrolyte, mechanical fracture of the brittle electrolyte phase under electro-chemo-mechanical loading, and the nucleation and propagation of lithium filaments that can short-circuit the device [4–6]. These processes are strongly coupled, as volume changes during cycling induce stress concentrations that promote decohesion and crack growth, which in turn exacerbate ionic transport heterogeneity and accelerate electrochemical degradation.

Over the past decade, numerous modeling strategies have been developed to describe these complex multiphysics phenomena. Continuum-level frameworks combining transport, mechanics, and electrochemistry have been employed to capture chemo-mechanical

stresses and their effect on interfacial failure [7,8]. At finer scales, discrete particle and microstructure-resolved simulations have provided insight into crack initiation and stress localization within polycrystalline electrolytes [9,10]. More recently, phase-field and cohesive-zone approaches have been introduced to represent fracture and interfacial decohesion in a thermodynamically consistent manner [11,12]. While these models have significantly advanced understanding, they often rely on homogenized or idealized microstructural descriptions, which can underestimate the impact of heterogeneity arising from processing-induced variability. Recent high-resolution experimental characterization, for example using synchrotron X-ray micro-computed tomography, has revealed pronounced microstructural heterogeneity in composite cathodes and pouch-scale all-solid-state batteries, underscoring the need for modeling frameworks that can account for spatial variability in material properties [13].

To bridge this gap, stochastic modeling has gained attention as a means of embedding microstructural uncertainty into continuum-level descriptions. By representing material properties as random fields, such approaches can capture the spatial variability of elasticity, strength,

[☆] This article is part of a Special issue entitled: 'Multiphysics Modeling' published in Advances in Engineering Software.

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and transport properties that govern local stress and failure [14]. Incorporating such variability is particularly relevant for ASSBs, where non-uniform contact, particle packing, and processing defects strongly influence electro-chemo-mechanical degradation pathways.

In this work, we introduce a stochastic framework for modeling multi-correlated random fields of material properties in solid electrolytes, with the aim of capturing heterogeneity-driven stress localization and interfacial degradation. The proposed methodology extends conventional continuum descriptions by embedding non-Gaussian, correlated property fluctuations constructed from a Gaussian translation field. Numerical simulations are presented to demonstrate how this approach enables realistic prediction of interface failure mechanisms in ASSBs.

2. Physics-based modeling

In this section, we present the continuum-level numerical model for simulating the response of the half-cell both in bulk and on material interfaces. Stochastic modeling of multiple correlated marginal random fields is presented.

2.1. Notation

We use italic symbols to denote scalar quantities (a) and bold symbols to denote vectors and tensors ($\mathbf{a}$ or $\mathbf{A}$). We denote vector inner products as $\mathbf{a} \cdot \mathbf{b}$, tensor–vector products as $\mathbf{A}\mathbf{b}$, and double-contraction between tensors as $\mathbf{A} : \mathbf{B}$. The symbol ∇ denotes the gradient operator in the reference configuration. $\dot{a}$ indicates a derivative in time. The L^2 -inner product on measure space X is $(a, b)_X$.¹ A comma in the subscript denotes partial differentiation, e.g. $a_{,bc}$ represents

$$a_{,bc} := \frac{\partial^2 a}{\partial b \partial c}.$$

The diamond subscript denotes a reference quantity, e.g. $c_\diamond$ and $T_\diamond$, depending on the context.

2.2. The initial–boundary value problem

This section summarizes an initial–boundary value problem (IBVP) for the solid-state battery incorporating electro-chemo-thermo-mechanical coupling. The IBVP is derived from variational principles following [15]. First, the governing equations resulting from the balance laws are

$$\rho c_p \dot{T} + \nabla \cdot \mathbf{h} - (r_{\text{int}} + r_{\text{ext}}) = 0, \quad \text{in } \Omega, \quad (1a)$$

$$\dot{c} + \nabla \cdot \mathbf{j} - m = 0, \quad \text{in } \Omega, \quad (1b)$$

$$\nabla \cdot \mathbf{i} = 0, \quad \text{in } \Omega, \quad (1c)$$

$$-\nabla \cdot \mathbf{P} - \mathbf{b} = 0, \quad \text{in } \Omega. \quad (1d)$$

The following are appropriate Neumann/Robin and Dirichlet boundary conditions

$$\mathbf{h} \cdot \mathbf{n} = h_n + h_{BV}, \quad \text{on } \partial\Omega, \quad (2a)$$

$$\mathbf{j} \cdot \mathbf{n} = j_n, \quad \text{on } \partial\Omega, \quad (2b)$$

$$\mathbf{i} \cdot \mathbf{n} = i_n + i_{BV}, \quad \text{on } \partial\Omega, \quad (2c)$$

$$\mathbf{P}\mathbf{n} = \mathbf{t}, \quad \text{on } \partial\Omega. \quad (2d)$$

The initial conditions for the temperature and the chemical concentration are trivial, i.e.

$$T = T_0, \quad \text{in } \Omega, \quad \text{at } t = 0, \quad (3a)$$

¹ The subscript X is dropped if the inner product is defined on the entire computational domain.

Table 1

Summary of relevant material properties in the IBVP for modeling the solid-state battery. Only macroscopic measurable properties are included. Other properties, e.g. the mobility M and the diffusional conductivity σ^d , can be inferred.

Name	Symbol	SI (derived) unit
Mass density	ρ	kg m^{-3}
Specific heat capacity	c_p	$\text{J kg}^{-1} \text{K}^{-1}$
Thermal conductivity	κ	$\text{J m}^{-1} \text{K}^{-1} \text{s}^{-1}$
Mass diffusivity	D	$\text{m}^2 \text{s}^{-1}$
Transference number	t_+	nondim.
Fugacity coefficients	f_+, f_-	nondim.
Ionic conductivity	σ^e	S m^{-1}
Charge transfer coefficient	α^e	nondim.
Swelling coefficient	α^s	nondim.
Molar volume	Ω	$\text{m}^3 \text{mol}^{-1}$
Thermal expansion coefficient	α^t	K^{-1}
Bulk modulus	λ	Pa
Shear modulus	G	Pa

$$c = c_0, \quad \text{in } \Omega, \quad \text{at } t = 0, \quad (3b)$$

where $T_0, c_0 \in \Omega \rightarrow \mathbb{R}_{>0}$ are the initial temperature and the initial chemical concentration, respectively.

The constitutive models are summarized below for convenience:

$$\text{internal heat flux: } \mathbf{h} = -\kappa_0 \nabla T, \quad (4a)$$

$$\text{reaction heat flux: } h_{BV} = i_{BV} \eta, \quad (4b)$$

$$\text{internal heat source: } r_{\text{int}} = \nabla \Phi \cdot \sigma_0^e \nabla \Phi, \quad (4c)$$

$$\text{internal mass flux: } \mathbf{j} = -M_0 \nabla \mu, \quad (4d)$$

$$\mu = \mu_\diamond + 2RT \ln \left(\frac{f_\pm c}{c_\diamond} \right) + \alpha^s \Omega p^m \quad (4e)$$

$$\text{internal current density: } \mathbf{i} = -\sigma_0^e \nabla \Phi - \frac{1}{F} \sigma_0^e (t_+ \nabla \mu_+ + t_- \nabla \mu_-), \quad (4f)$$

$$\text{reaction overpotential: } \eta = g(D) \|\Phi\| - U, \quad (4g)$$

$$\text{reaction current density: } i_{BV} = 2i_0 \sinh \left(\frac{\alpha^e F}{RT} \eta \right), \quad (4h)$$

$$\text{deformation gradients: } \mathbf{F}^s = \mathbf{J}^{\frac{1}{3}} \mathbf{I}, \quad \mathbf{J}^s = 1 + \alpha^s \Omega (c - c_\diamond), \quad (4i)$$

$$\mathbf{F}^t = \mathbf{J}^{t \frac{1}{3}} \mathbf{I}, \quad \mathbf{J}^t = 1 + \alpha^t (T - T_\diamond), \quad (4j)$$

$$\text{the PK-1 stress: } \mathbf{P} = [\lambda \ln(I_3) \mathbf{F}^{m-T} + G (\mathbf{F}^m - \mathbf{F}^{m-T})] (\mathbf{F}^s \mathbf{F}^t)^{-1}. \quad (4k)$$

Eqs. (1), (2), (3) & (4) completely define the IBVP up to a specific choice of geometry, material properties, and external loadings. In the reaction overpotential, $g(D)$ is a scalar degradation function that couples interfacial mechanical damage into the local charge-transfer kinetics; it is defined in Eq. (8) and its role is discussed in Section 2.3. Table 1 summarizes the required material properties.

2.3. Interface degradation

In addition to chemical reactions described by the Butler–Volmer Eq. (4), we introduce a traction–separation relation at material interfaces. The traction–separation relation is adapted from the one originally proposed in [16]. The traction–separation law is equipped with two criteria for predicting the propagation of interface decohesion characterized by a damage indicator D .

The traction–separation relation is defined in the local coordinates ($\mathbf{e}_n, \mathbf{e}_s, \mathbf{e}_t$) with the convention that $\mathbf{e}_n$ is the outward normal of the interface, and that $\mathbf{e}_s$ and $\mathbf{e}_t$ span the tangent plane. Together, ($\mathbf{e}_n, \mathbf{e}_s, \mathbf{e}_t$) form an orthonormal basis. The displacement jump and traction in the global Cartesian coordinates are projected onto this local basis as

$$\llbracket \mathbf{u} \rrbracket = \llbracket \mathbf{u} \rrbracket_n \mathbf{e}_n + \llbracket \mathbf{u} \rrbracket_s \mathbf{e}_s + \llbracket \mathbf{u} \rrbracket_t \mathbf{e}_t, \quad (5a)$$

$$\llbracket \mathbf{u} \rrbracket_n = \llbracket \mathbf{u} \rrbracket \cdot \mathbf{e}_n, \quad (5b)$$

$$\llbracket u \rrbracket_s = \llbracket u \rrbracket \cdot e_s, \quad (5c)$$

$$\llbracket u \rrbracket_t = \llbracket u \rrbracket \cdot e_t. \quad (5d)$$

$$t = t_n e_n + t_s e_s + t_t e_t, \quad (6a)$$

$$t_n = t \cdot e_n, \quad (6b)$$

$$t_s = t \cdot e_s, \quad (6c)$$

$$t_t = t \cdot e_t. \quad (6d)$$

With a view towards preventing interface penetration, the displacement jump is additively decomposed into an active part subject to damage softening and an inactive part which remains intact, i.e.,

$$\llbracket u \rrbracket = \llbracket u \rrbracket^{(A)} + \llbracket u \rrbracket^{(I)}, \quad (7a)$$

$$\llbracket u \rrbracket^{(A)} = \langle \llbracket u \rrbracket_n \rangle e_n + \llbracket u \rrbracket_s e_s + \llbracket u \rrbracket_t e_t, \quad (7b)$$

$$\llbracket u \rrbracket^{(I)} = (\llbracket u \rrbracket_n - \langle \llbracket u \rrbracket_n \rangle) e_n, \quad (7c)$$

where $\langle \cdot \rangle$ is the standard Macaulay bracket. The active part is softened by a scalar degradation function

$$g(D) = 1 - D, \quad (8)$$

where $D \in [0, 1]$ is the interfacial damage index. The traction-separation law then takes the following general form

$$t = \hat{t}(D, \llbracket u \rrbracket) = g(D) K \llbracket u \rrbracket^{(A)} + K \llbracket u \rrbracket^{(I)}, \quad (9)$$

where K is the stiffness of the interface. In the numerical implementation, the analytical $g(D)$ is regularized (both in the cohesive traction of Eq. (9) and in the reaction overpotential of the constitutive list (4)) so that it takes a small positive value $\varepsilon \ll 1$ at $D = 1$ rather than exactly zero, preventing singular behavior of the coupled solver as damage saturates. The regularization is chosen so that the analytical relation is recovered in the $\varepsilon \rightarrow 0$ limit. The onset of softening (damage initiation) is governed by a mode-mixity-dependent strength envelope defined as

$$\phi^t = \|\mathbf{t}\|^2 - \frac{(1 + \beta^2)N^2 S^2}{\beta^2 N^2 + S^2} \leq 0, \quad \dot{D} \geq 0, \quad \phi^t \dot{D} = 0, \quad (10a)$$

$$\beta = \frac{\sqrt{\llbracket u \rrbracket_s^2 + \llbracket u \rrbracket_t^2}}{\langle \llbracket u \rrbracket_n \rangle}. \quad (10b)$$

where $\beta \in \mathbb{R}_+$ is the mode mixity factor. When the opening is purely mode I, i.e., $\llbracket u \rrbracket_n > 0$ and $\llbracket u \rrbracket_s = \llbracket u \rrbracket_t = 0$, the mode mixity factor $\beta = 0$. N and S are the interface strength in the normal and tangential directions, respectively.

A mode-mixity-dependent strength envelope is necessary in the present context because cathode–electrolyte interfaces in composite cathodes are loaded under arbitrary combinations of normal opening and tangential sliding rather than pure mode I. During electrochemical cycling, the volumetric swelling of NMC811 particles produces a complex multi-axial traction state on the cathode–electrolyte interface that depends on the local geometry of neighboring particles (see, e.g., Fig. 9(a)). A pure mode-I (normal-stress only) initiation criterion would systematically overpredict the damage threshold for interfaces whose primary loading is tangential, and would fail to recover the experimentally observed sensitivity of interface failure to particle arrangement. The envelope in Eq. (10a) interpolates smoothly between the pure-normal limit $\|\mathbf{t}\| = N$ at $\beta = 0$ and the pure-tangential limit $\|\mathbf{t}\| = S$ as $\beta \rightarrow \infty$, recovering the expected single-mode behavior while providing a physically motivated, continuously differentiable surface for arbitrary mode mixities. Before the onset of softening, i.e., $D = 0$, the strength surface (10a) can be mapped to the displacement space using the traction–separation relation (9) as

$$\phi^t = \|\llbracket u \rrbracket\| - \llbracket u \rrbracket_c, \quad \llbracket u \rrbracket_c = \frac{\sqrt{(1 + \beta^2)\llbracket u \rrbracket_n \llbracket u \rrbracket_{s,c}}}{\sqrt{\beta^2 \llbracket u \rrbracket_n^2 + \llbracket u \rrbracket_{s,c}^2}}, \quad \llbracket u \rrbracket_{n,c} = \frac{N}{K}, \quad \llbracket u \rrbracket_{s,c} = \frac{S}{K}, \quad (11)$$

where $\llbracket u \rrbracket_c$ is the effective critical displacement jump corresponding to the onset of softening, and $\llbracket u \rrbracket_{n,c}$ and $\llbracket u \rrbracket_{s,c}$ are the critical normal and tangential jumps in the corresponding pure modes. Two propagation criteria are considered in this work: the power-law criterion for transgranular crack propagation and the Benzeggagh–Kenane criterion for intergranular decohesion propagation. The mixed-mode critical energy release rates are defined as

$$\mathcal{G}_c = \begin{cases} (1 + \beta^2) \left[\left(\frac{1}{G_{I,c}} \right)^n + \left(\frac{\beta^2}{G_{II,c}} \right)^n \right]^{-1/n}, & \text{power-law} \\ G_{I,c} + (G_{II,c} - G_{I,c}) \left(\frac{\beta^2}{1 + \beta^2} \right)^n, & \text{Benzeggagh–Kenane} \end{cases} \quad (12)$$

where $G_{I,c}$ and $G_{II,c}$ are the mode-I and mode-II critical energy release rates, respectively, and n is the (dimensionless) exponent controlling the rate at which $\mathcal{G}_c$ transitions from $G_{I,c}$ to $G_{II,c}$ as the loading becomes more shear-dominated. Fig. 1 illustrates the variation of $\mathcal{G}_c$ with β for representative values of $G_{II,c}/G_{I,c}$ and n , for both criteria. Both forms recover $\mathcal{G}_c = G_{I,c}$ in pure mode I ($\beta = 0$) and approach $G_{II,c}$ as $\beta \rightarrow \infty$; larger n produces a sharper transition between the two limits.

The softening part of the bilinear traction–separation law can be derived by solving the following system of equations:

$$\|\hat{\mathbf{t}}\| = K \llbracket u \rrbracket_c \frac{\llbracket u \rrbracket_f - \|\llbracket u \rrbracket\|}{\llbracket u \rrbracket_f - \llbracket u \rrbracket_c}, \quad (13a)$$

$$\frac{1}{2} K \llbracket u \rrbracket_c \llbracket u \rrbracket_f = \mathcal{G}_c. \quad (13b)$$

(13a) is the linear-softening branch of the bilinear traction–separation curve, corresponding to elastic unloading with the degraded stiffness $g(D)K$. (13b) enforces that the correct amount of surface energy is released upon complete failure: $\llbracket u \rrbracket_f$ denotes the effective failure displacement jump, i.e., the magnitude of $\llbracket u \rrbracket$ at which the interface is fully decohered ($D = 1$) and the active cohesive stiffness has degraded to zero (analytically) or to the small residual value εK retained by the numerical regularization. Geometrically, $\llbracket u \rrbracket_c$ and $\llbracket u \rrbracket_f$ are the two endpoints of the linear softening branch in the bilinear traction–separation diagram, and the area $\frac{1}{2} K \llbracket u \rrbracket_c \llbracket u \rrbracket_f$ enclosed by the diagram equals $\mathcal{G}_c$ as enforced by (13b). Solving the system of equations yields the expression for damage

$$D = \hat{D}(\|\llbracket u \rrbracket\|) = \begin{cases} 0, & \|\llbracket u \rrbracket\| \in [0, \llbracket u \rrbracket_c) \\ \frac{\llbracket u \rrbracket_f (\|\llbracket u \rrbracket\| - \llbracket u \rrbracket_c)}{\llbracket u \rrbracket_f (\llbracket u \rrbracket_f - \llbracket u \rrbracket_c)}, & \|\llbracket u \rrbracket\| \in [\llbracket u \rrbracket_c, \llbracket u \rrbracket_f) \\ 1, & \|\llbracket u \rrbracket\| \in [\llbracket u \rrbracket_f, \infty) \end{cases} \quad (14)$$

$$\llbracket u \rrbracket_f = \frac{2\mathcal{G}_c}{K \llbracket u \rrbracket_c}. \quad (15)$$

To close the electro-chemo-mechanical coupling at the interface, the same damage variable D that softens the cohesive traction also degrades the electric-potential jump driving the Butler–Volmer charge-transfer reaction through the $g(D)$ factor in the reaction overpotential of (4). As damage accumulates ($D \rightarrow 1$), the mechanical load-carrying capacity of the interface degrades through the softened cohesive traction in Eq. (9), and the electric-potential-jump contribution to the interfacial overpotential is scaled by $g(D)$, feeding damage back into the local Butler–Volmer reaction current. Because η retains the equilibrium-potential contribution $-U$, the reaction current is not driven identically to zero by damage alone; its evolution with D depends on the local relative magnitudes of $\|\Phi\|$ and U . In the operating regime of the simulations presented in Section 5, where $\|\Phi\|$ dominates during charging, scaling $\|\Phi\|$ by $g(D)$ yields a progressive reduction of the local charge transfer as decohesion accumulates. This is the two-way electro-chemo-mechanical coupling at the interface: bulk electrochemistry drives mechanical deformation through swelling (Eq. (4)), the resulting interfacial stress concentration produces damage D through

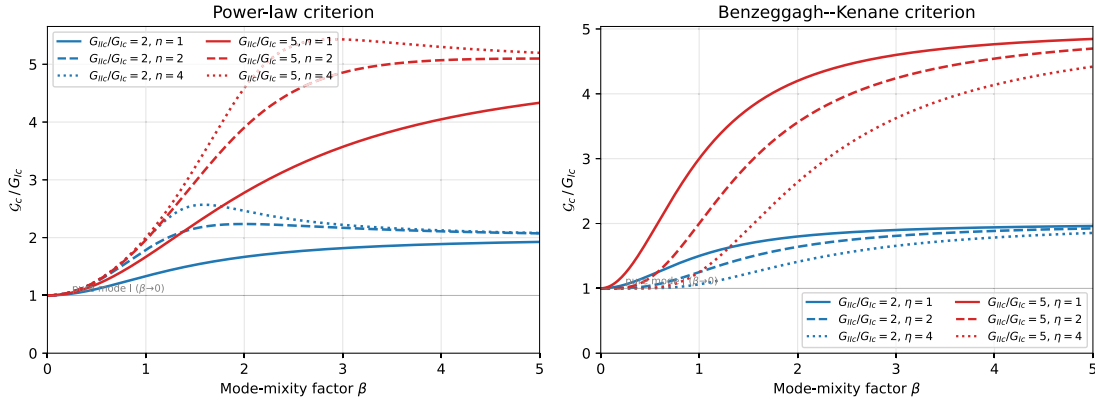


Fig. 1. Variation of the mixed-mode critical energy release rate G_c (normalized by $G_{I,c}$) with the mode-mixity factor β , for the power-law criterion (left) and the Benzeggagh–Kenane criterion (right). Two ratios of mode-II to mode-I toughness ($G_{II,c}/G_{I,c} = 2$ and 5) and three values of the exponent n are shown to illustrate the role of these parameters.

Eqs. (10a)–(14), and the accumulated damage in turn degrades the local interfacial charge transfer through Eq. (4).

We emphasize that, in the present formulation, damage feedback is restricted to interfacial transport. The bulk ionic conductivity σ^e , ion diffusivity D , and mobility M of the electrolyte phase are *not* modified by interfacial damage; only their stochastic spatial variability is captured through the random fields described in Section 3. This modeling choice is motivated by two considerations. First, the present continuum description does not resolve bulk cracks: the only damage variable is the interfacial D defined on the cathode–electrolyte sidesets, so introducing a bulk-damage–transport coupling would require an additional bulk damage field (e.g., a phase-field crack representation), which lies beyond the present scope. Second, in NMC/LPSCI composite cathodes under stress cycling, the cathode–electrolyte interface, rather than the bulk electrolyte, has been consistently identified as the dominant site of impedance growth and capacity fade; isolating the interfacial pathway therefore provides a physically motivated test of the mechanical-damage hypothesis pursued in this work.

Combining (9) and (14) yields the implicit system for updating the interface traction t and damage D . The residuals of the implicit system can be written as

$$r_t(t, D; \|\mathbf{u}\|) = t - \hat{t}(D, \|\mathbf{u}\|) \quad (16a)$$

$$r_D(D; \|\mathbf{u}\|) = v\dot{D} - [\hat{D}(\|\mathbf{u}\|) - D] \quad (16b)$$

$$\text{subject to } \dot{D} \geq 0 \quad (16c)$$

where an artificial viscosity v is added to regularize the softening process. Implicitly updating the system (16) is numerically challenging due to the loss of stability when damage exceeds a certain threshold. To improve numerical stability, we modify the system based on the observation that the traction subproblem, i.e., r_t , is always stable in the direction of δt , and, similarly, the damage subproblem r_D is always stable in the direction of δD . The modified implicit system becomes

$$r_t(t, D; \|\mathbf{u}\|) = t - \hat{t}(D, \|\mathbf{u}\|) \quad (17a)$$

$$r_D(D; \|\bar{\mathbf{u}}\|) = v\dot{D} - [\hat{D}(\|\bar{\mathbf{u}}\|) - D] \quad (17b)$$

$$\text{subject to } \dot{D} \geq 0 \quad (17c)$$

where $\bar{\mathbf{u}}$ denotes the previously converged displacements. The above modification leads to a stable multi-step update algorithm of the traction–separation relation using fixed-point iteration. At each step, the damage subproblem, i.e., (17b) and (17c), is first solved to update the damage, and the traction is then updated using (17a) with the updated damage. The fixed-point iteration can be repeated until t and D converge.

3. Stochastic modeling of correlated material-property fields

The degradation of solid–solid interfaces in all-solid-state batteries is closely tied to localized stress concentrations that arise under electrochemical cycling. As discussed in Section 2.3, decohesion is driven by mechanical separation laws. However, continuum-level homogenizations of the microstructure (e.g., treating electrolyte particles and conductive additives as lumped entities) often fail to capture such stress localization, which leads to non-conservative predictions of interfacial failure.

To address this limitation while retaining computational tractability, we enrich the continuum description by embedding spatial variability into the constitutive parameters of the electrolyte. This is accomplished by representing the material properties as realizations of random fields that encode stochastic features of the manufacturing process. Concretely, we introduce a vector-valued field

$$\{\mathbf{P}(\mathbf{x}) = (P_1(\mathbf{x}), P_2(\mathbf{x}), P_3(\mathbf{x})), \mathbf{x} \in \Omega\}, \quad (18)$$

defined over a probability space (Θ, Σ, P) and taking values in $\mathbb{R}_{>0}^3$. The three components are intended as surrogate representations of the spatially heterogeneous elastic modulus $E(\mathbf{x})$, ionic conductivity $\sigma^i(\mathbf{x})$, and ion diffusivity $D(\mathbf{x})$. Since these quantities are strictly positive, $\mathbf{P}(\mathbf{x})$ cannot in general be modeled as a Gaussian field. Instead, we construct $\mathbf{P}$ through a nonlinear transformation of an auxiliary Gaussian field,

$$\mathbf{P}(\mathbf{x}) = \mathcal{T}(\Xi(\mathbf{x}), \mathbf{x}), \quad \mathbf{x} \in \Omega, \quad (19)$$

where Ξ is a centered Gaussian field in $\mathbb{R}^3$ and $\mathcal{T}$ is a measurable mapping chosen so that $\mathbf{P}$ exhibits prescribed one-point distributions. This approach, which follows a translation-field methodology [17], provides a reduced parametrization while preserving essential non-Gaussian statistics.

We enforce the marginal distributions $\{f_x\}_{x \in \Omega}$ by prescribing

$$P_{P(x)}(dp) = f_x(p) dp, \quad p \in \mathbb{R}^3, \quad (20)$$

with $dp = dp_1 dp_2 dp_3$. For simplicity, we assume spatial stationarity of the hyperparameters and write $f_x(\cdot) = f(\cdot; \mathbf{w})$. Guided by the maximum entropy principle [18,19], Gamma laws are selected as marginals, ensuring positivity and finite variance. Thus, for $i \in \{1, 2, 3\}$,

$$P_{P_i(x)}(dp_i) = f_G(p_i; (\underline{p}_i, \delta_i)) dp_i, \quad (21)$$

where f_G denotes the Gamma density with mean $\underline{p}_i$ and coefficient of variation δ_i .

Because only the marginals are specified, the construction of the joint law requires additional modeling. We adopt a correlation-based copula formulation: letting ρ_{ij} denote the Pearson correlation between

$P_i(\mathbf{x})$ and $P_j(\mathbf{x})$, the transformation is expressed through the Gaussian copula,

$$P_i(\mathbf{x}) = F_G^{-1}(\Phi(Y_i(\mathbf{x})); (\underline{p}_i, \delta_i)), \quad (22)$$

where $\mathbf{Y}(\mathbf{x})$ is a centered Gaussian vector with covariance matrix $C_Y = [\rho_{ij}]$ and Φ is the Gaussian CDF. Using the Cholesky factorization $C_Y = L_Y^\top L_Y$, one obtains

$$\mathbf{Y}(\mathbf{x}) = L_Y^\top \Xi(\mathbf{x}), \quad (23)$$

which completes the definition of $\mathcal{T}$.

The Gaussian driver field Ξ is assumed stationary on Ω , and is fully specified by its covariance function $R_\Xi(\tau) = \mathbb{E}\{\Xi(\mathbf{x} + \tau) \otimes \Xi(\mathbf{x})\}$. Realizations are generated using truncated Karhunen–Loève expansions,

$$\Xi_i(\mathbf{x}) = \sum_{k=1}^{q_i} \sqrt{\lambda_k} \eta_k \varphi_k(\mathbf{x}), \quad (24)$$

where $\{\eta_k\}$ are independent Gaussian variables, and (λ_k, φ_k) are eigenpairs of the covariance operator, i.e., solutions of the Fredholm integral equation

$$\int_{\Omega} R(\mathbf{x}, \mathbf{x}') \varphi_k(\mathbf{x}') d\mathbf{x}' = \lambda_k \varphi_k(\mathbf{x}), \quad \mathbf{x} \in \Omega. \quad (25)$$

Approximate solutions are obtained through Galerkin projection [20, 21], leading to a generalized eigenproblem involving the kernel matrix and the L^2 mass matrix. The truncation order q_i is selected by enforcing a tolerance on the residual variance,

$$\varepsilon_{\text{KL}}(q) = 1 - \frac{\sum_{k=1}^q \lambda_k}{\sum_{k=1}^{N_{\text{dof}}} \lambda_k} \leq \varepsilon_0. \quad (26)$$

In summary, the constructed field $\mathbf{P}(\mathbf{x})$ satisfies: (i) positivity almost surely, (ii) finite second-order moments, and (iii) constant mean values $\underline{p} = (\underline{p}_1, \underline{p}_2, \underline{p}_3)$. The framework enables efficient simulation of spatially correlated, non-Gaussian heterogeneity in electrolyte properties while retaining a continuum representation suitable for large-scale battery models.

4. Experimental methods (Half-cell cycling)

The overall half-cell architecture and the multi-phase composition of the composite cathode are illustrated schematically in Fig. 2. The cell consists of a $\text{Li}_{0.5}\text{In}$ alloy anode and a composite cathode separated by a pelletized LPSCI solid electrolyte. The fabricated composite cathode contains three phases: NMC811 active material (80 wt%), LPSCI solid electrolyte (17 wt%), and VGCF conductive additive (3 wt%), each of which contributes to a distinct transport pathway (Li^+ storage/insertion, ionic conduction, and electronic conduction, respectively). During charging, Li^+ ions are extracted from the NMC811 particles, transported through the LPSCI phase (both within the composite cathode and across the separator), and reduced at the $\text{Li}_{0.5}\text{In}$ anode. In the numerical model of Section 5, the composite cathode is represented as NMC811 particles (80 wt%) embedded in a solid-electrolyte matrix (the combined LPSCI + VGCF phase, 20 wt%); the effective properties assigned to the matrix (electrical conductivity, ionic diffusivity, elasticity) already incorporate the electronic-conduction contribution of VGCF.

Composite cathodes were prepared using lithium nickel cobalt manganese oxide (NMC811), lithium phosphorus sulfur chloride (LPSCI) solid electrolyte, and vapor-grown carbon fiber (VGCF) as a conductive additive. The powders were homogenized prior to cell assembly. All-solid-state half-cells were assembled using a pelletized LPSCI solid electrolyte and a $\text{Li}_{0.5}\text{In}$ alloy anode. Cell fabrication was carried out in a uniaxial die under controlled stack pressure. Galvanostatic cycling was performed at room temperature under constant pressure. After initial formation cycles at low rate, cells were cycled within a voltage window corresponding to approximately 2–3.65 V versus $\text{Li}_{0.5}\text{In}$ (equivalent to 2.65–4.3 V versus Li/Li^+).

The experimental cycling data presented in this work are used to calibrate interfacial degradation parameters and to validate trends in capacity fade and depth of discharge predicted by the stochastic continuum simulations.

5. A half-cell model

In this section, a comprehensive half-cell model of an all-solid-state battery is presented. The half-cell consists of NMC811 cathode active material, LPSCI solid electrolyte, and VGCF as the conductive additive. Interface decohesion parameters are calibrated against experimental cycling data. Contour plots of the maximum principal stress and ion concentration at the end of the simulated cycling are then presented to provide insight into strongly localized interface degradation.

5.1. Software implementation

All numerical simulations reported in this work were performed using EEL, an open-source multiphysics finite-element code for electro-chemo-thermo-mechanical modeling of solid-state batteries developed at Argonne National Laboratory [15,22]. EEL is implemented in C++ as a downstream application of the MOOSE framework [23], and inherits from it the underlying finite-element and parallel-computing infrastructure: the mesh, degree-of-freedom management, and assembly routines are provided by libMesh, and the linear/nonlinear solvers and eigen-solvers by PETSc [24] and SLEPc [25], respectively. All simulations use double-precision arithmetic and MPI-based domain decomposition for parallel execution.

The initial-boundary value problem of Section 2.2 is discretized in space using first-order Lagrange (P^1) polynomials on a conforming quadrilateral (2D) or hexahedral (3D) mesh; the resulting semidiscrete Galerkin form is integrated in time using the backward Euler method. Material interfaces (Section 2.3) are implemented by disconnecting element pairs across pre-defined sidesets and re-coupling them through an interior-penalty discontinuous-Galerkin formulation, following the interface treatment described in [15]; the cohesive traction–separation law is evaluated as a MOOSE InterfaceMaterial, and the interface degradation described in Section 2.3 is implemented directly on top of this facility. Consistent Jacobians for the coupled Newton system are computed automatically through MOOSE’s forward-mode automatic differentiation (AD) facility, which alleviates the need for hand-coded tangent operators.

The stochastic modeling infrastructure of Section 3 is provided by a standalone utility that (i) assembles the Fredholm eigenvalue problem (25) using libMesh’s finite-element assembly, (ii) solves it with SLEPc’s Krylov–Schur algorithm to obtain the KL eigenpairs (retained up to the truncation tolerance of Eq. (26)), and (iii) samples correlated Gamma-distributed random fields via the Gaussian-copula transformation of Eq. (22). The resulting property fields are written to an Exodus mesh file that is then read as spatially varying material properties by the coupled electro-chemo-mechanical simulation.

EEL, the underlying open-source multiphysics framework used in this work, is publicly available under the LGPL-3.0 license at <https://github.com/hugary1995/eel> (documentation: <https://hugary1995.github.io/eel/>). A detailed technical report describing the numerical framework, verification against analytical benchmarks, and additional application examples is available as ANL report ANL-22/67 [22].

5.2. Continuum representation of the solid electrolyte

Consider a two-dimensional domain $\Omega = [0, 0.018 \text{ mm}] \times [0, 0.018 \text{ mm}]$ representing a half-cell consisting of random close packing of circular particles representing the solid electrolyte. Strongly correlated random fields can be generated following Section 3 to provide statistically equivalent continuum representation of the electrolyte’s Young’s modulus, ionic conductivity, and ion diffusivity.

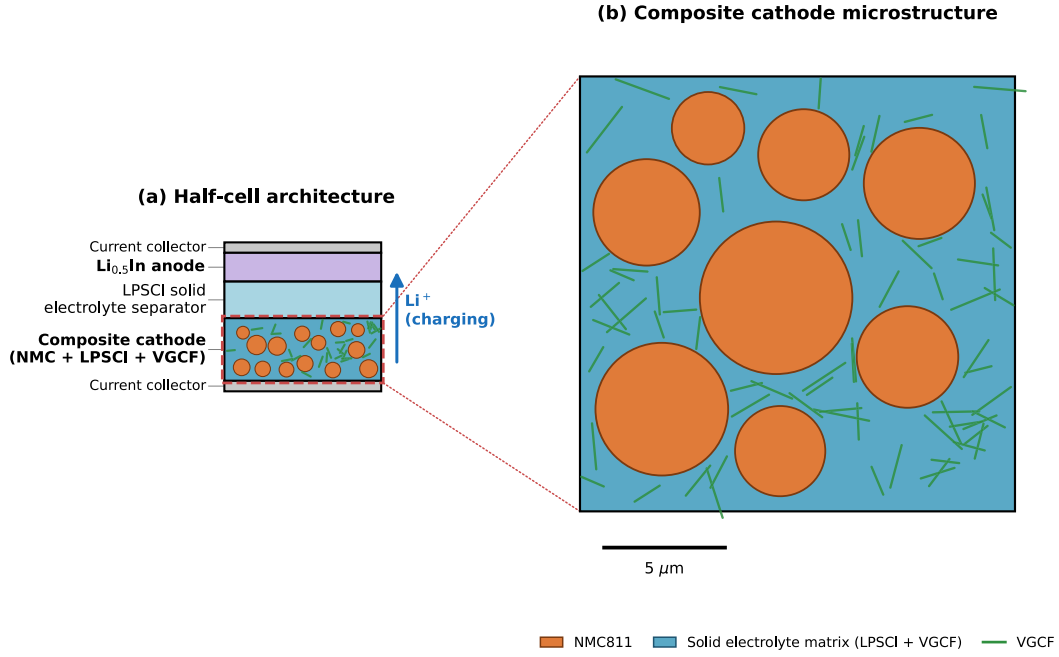


Fig. 2. Schematic of the all-solid-state half-cell studied in this work. (a) Layer-by-layer architecture: current collectors sandwich a $\text{Li}_{0.5}\text{In}$ alloy anode, a pelletized LPSCI solid electrolyte separator, and the composite cathode. During charging, Li^+ migrates from the composite cathode through the LPSCI separator to the anode (blue arrow). The dashed red box highlights the composite cathode region, expanded in (b). (b) Representative $18 \times 18 \mu\text{m}^2$ RVE of the composite cathode showing NMC811 active-material particles (orange), LPSCI solid-electrolyte particles (blue), and VGCF conductive-additive fibers (black lines). Visual proportions are illustrative and do not reflect area or volume fractions.

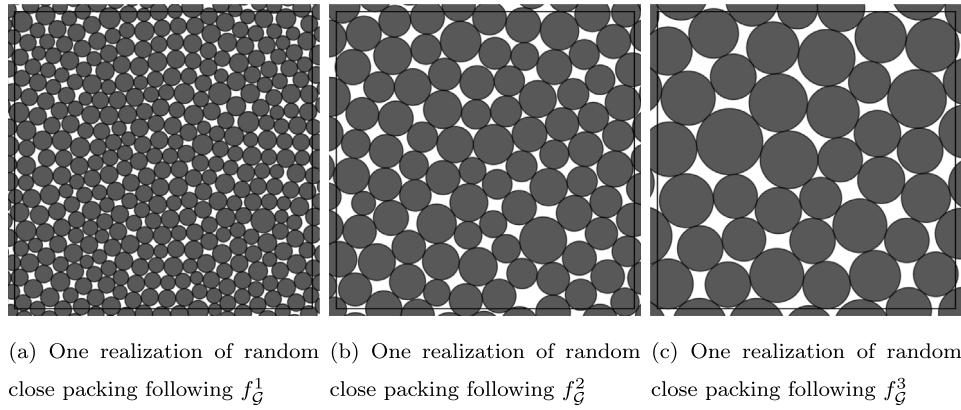


Fig. 3. Random close packings of solid electrolytes with different radius distributions.

The particle radius is assumed to follow a Gamma distribution with mean $\bar{r}$ and coefficient of variation $\delta = 0.1$. Three distributions are considered for demonstration purposes: $f_G^1(r; 0.5 \mu\text{m}, 0.1)$, $f_G^2(r; 1 \mu\text{m}, 0.1)$, and $f_G^3(r; 1.5 \mu\text{m}, 0.1)$. Monodisperse random close packings are generated using the hierarchical packing algorithm described in [26]. One realization from each radius distribution is illustrated in Fig. 3.

Each particle is assumed to have a homogeneous Young's modulus following Gamma distribution $f_G(E; \bar{E} = 1 \times 10^4 \text{ MPa}, \delta_E = 0.1)$. The normalized property field $\tilde{E}(\mathbf{x})$, for $\mathbf{x} \in \Omega$ can be written as

$$\tilde{E}(\mathbf{x}) = \sum_{i=1}^{N_c} \tilde{E}_i \mathbb{1}_i(\mathbf{x}), \quad (27)$$

where N_c is the number of particles, $\tilde{E}_i$ is the normalized Young's modulus of particle realization i (see e.g. (18)), and $\mathbb{1}_i$ is the indicator of particle i . Assuming the spatial covariance is stationary and isotropic,

the following empirical estimator can be used

$$\hat{R}(h) = \frac{1}{|\Omega|} \sum_{i,j} \tilde{E}_i \tilde{E}_j |V_i \cap (V_j - h)| - \hat{\bar{E}}^2, \quad \hat{\bar{E}} = \frac{1}{|\Omega|} \sum_i \tilde{E}_i |V_i|. \quad (28)$$

The estimate for each realization shown in Fig. 3 is presented in Fig. 4.

The estimated covariance functions define the Karhunen-Loève expansion of the underlying Gaussian fields. Resorting to the discretization of the Fredholm integral equation of the second kind, i.e. (25), on a Cartesian grid with spacing $h^e = 0.36 \mu\text{m}$ (a 50×50 -element P^1 -Lagrange grid comprising 51×51 nodes, i.e., $N_{\text{dof}} = 2601$), coefficients of (24) are determined with a truncation error of 1% (see (26)). For the three packing realizations shown in Fig. 3, applying this criterion retains $q_1 \approx 420$, $q_2 \approx 115$, and $q_3 \approx 56$ modes, respectively; the decreasing trend reflects the longer effective correlation length of the underlying property field as the mean particle radius increases. These values reflect a squared-exponential fit to the empirical covariance functions of

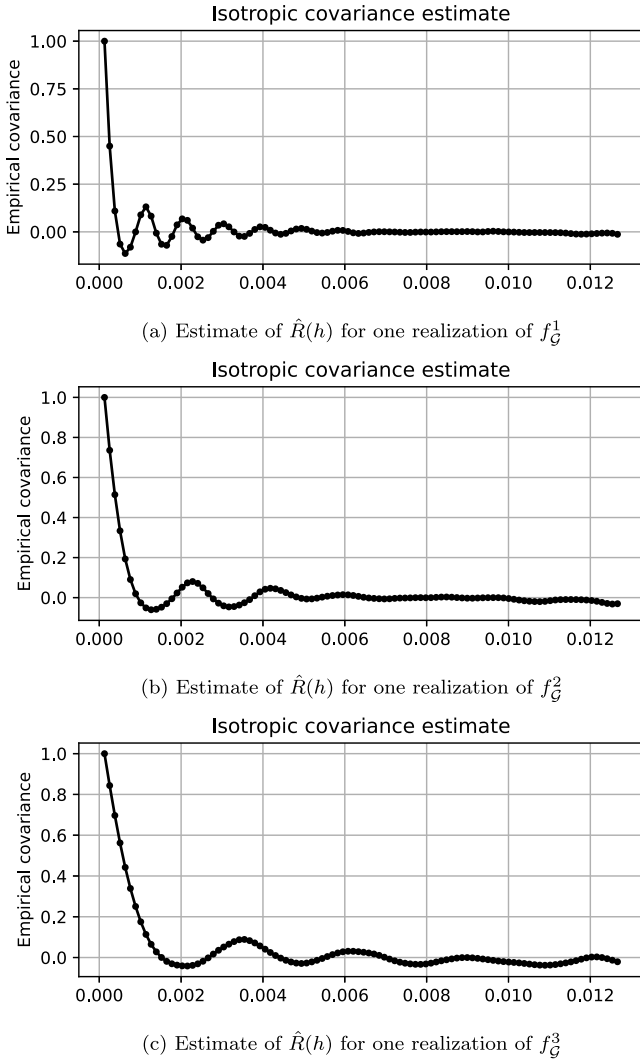


Fig. 4. Empirical estimates of the isotropic covariance function of the Young's modulus for the realizations shown in Fig. 3.

Fig. 4; the precise counts vary by $\mathcal{O}(10\%)$ depending on the kernel model. The corresponding cumulative eigenvalue spectra are shown in Fig. 5. Realization of the Gaussian random fields $\tilde{E}(\mathbf{x})$ is then simplified to the sampling of the independent normalized Gaussian random variables $\{\eta_k\}$. The Gamma random fields $E(\mathbf{x})$ can be obtained by evaluating the nonlinear transformation defined by (22). For demonstration purposes, one realization of $E(\mathbf{x})$ for each combination of hyperparameters is shown in Fig. 6. Similar random fields are generated for the ionic conductivity and the ion diffusivity. Note that the point-wise Pearson-type correlation can be controlled in the sense defined by Eqs. (22) and (23). However, in this example, the three random fields are assumed to be mutually uncorrelated, i.e., $\rho_{ij} = \delta_{ij}$.

Tortuosity of the random fields is defined as the domain integral of the localized geometric deviation with respect to a reference direction, i.e.

$$\tau = \int_{\Omega} \left(1 - \frac{\mathbf{j} \cdot \mathbf{d}}{\|\mathbf{j}\|} \right) dV, \quad (29)$$

where $\mathbf{j}$ is the diffusion flux (i.e., either current density or mass flux, depending on the context), and $\mathbf{d}$ is the unit reference flux in a continuous homogeneous medium. Using this definition, the converged moments of the tortuosity are summarized in Table 2.

Table 2

Summary of the resulting tortuosity (29) and the maximum local tortuosity given by the Gamma random fields with different hyperparameters.

Mean particle radius (μm)	Tortuosity ($\times 10^{-3}$)	Max. local tortuosity ($\times 10^{-3}$)
0.5	0.254 ± 0.0159	3.73 ± 0.764
1	0.461 ± 0.0328	6.79 ± 2.15
1.5	0.494 ± 0.0159	6.96 ± 1.78

Given the continuum representation of electrolyte properties, non-overlapping NMC811 cathode particles are randomly placed within the RVE. The radius of the cathode particles is modeled as a random variable with distribution $f_G(r; 3\mu\text{m}, 0.1)$. Each NMC particle is treated as a homogeneous elastic-swelling continuum body with lumped effective properties; the internal polycrystalline architecture of NMC811 secondary particles (nanoscale primary crystallites of order 100 nm bonded through grain boundaries, e.g., [10]) is not explicitly resolved at the RVE scale considered here. Table 3 summarizes the relevant length scales in the present model.

In the following calculations, the NMC811 cathode particles are assumed to be homogeneous: electrical conductivity $\sigma^e = 0.1 \text{ S cm}^{-1}$, ionic diffusivity $D = 5 \times 10^{-5} \text{ mm}^2 \text{ s}^{-1}$, Young's modulus $E = 1 \times 10^5 \text{ MPa}$, Poisson's ratio $\nu = 0.3$, and molar volume (per mole of Li in the host lattice) $\Omega = 10 \text{ cm}^3 \text{ mol}^{-1}$. Mean values of the LPSCl solid electrolyte medium are: electrical conductivity $\sigma^e = 1 \times 10^{-9} \text{ S cm}^{-1}$, ionic diffusivity $D = 1 \times 10^{-4} \text{ mm}^2 \text{ s}^{-1}$, Young's modulus $E = 1 \times 10^4 \text{ MPa}$, Poisson's ratio $\nu = 0.25$, and molar volume (per mole of Li in the host lattice) $\Omega = 4 \text{ cm}^3 \text{ mol}^{-1}$.

The cathode–electrolyte interface is assumed to be rigid at its intact state, with homogeneous properties: interfacial normal stiffness $E = 1 \times 10^7 \text{ MPa}$ and shear stiffness $G = 5 \times 10^6 \text{ MPa}$. The reference traction $T_n^0 = 2200 \text{ MPa}$ and the degradation power-law exponent $n = 6$ are calibrated to match experimental data. The interface exchange current is $i_0 = 1 \times 10^{-4} \text{ mA mm}^{-2}$, with open-circuit potential

$$U = 5.5826 - 6.9922\alpha + 7.1062\alpha^2 - 5.4549 \times 10^{-5} \exp(124.23\alpha - 114.2593) - 2.5947\alpha^3, \quad (30)$$

with the transfer degree α defined as the average Hermite interpolation of the lithium saturation in the neighboring cathode and electrolyte.

The RVE is fully fixed at all external boundaries. A constant current is applied on one side of the RVE to simulate the charging process. The charging process terminates when the electric potential $\Delta\Phi$ across the cell is above 3.65 V. The discharging process immediately follows the charging process and terminates when the electric potential is below 2 V. The charging–discharging cycle is repeated until the half-cell no longer charges or discharges. Numerically, the final abrupt failure is indicated by an increase in electric potential during discharging or a decrease in electric potential during charging.

Before presenting the cycling results, we clarify the degradation mechanism resolved by the model. Cycle-to-cycle capacity fade in the present simulations arises from a single, physically explicit source: the accumulation of interfacial damage D at cathode–electrolyte contacts through the traction–separation law of Section 2.3, which is monotonically increasing in time ($\dot{D} \geq 0$). As D grows, the local electric-potential jump driving the Butler–Volmer reaction is scaled by $g(D)$ through the reaction overpotential in the constitutive list (4). In the operating regime of these simulations (where $\|\Phi\|$ dominates the overpotential during charging), this scaling produces a progressive reduction of the local ion-transfer capacity and, cumulatively, of both the specific differential capacity (Fig. 7) and the depth of discharge (Fig. 8) at higher cycle counts. Physically, this reduction reflects a progressive loss of accessible charge-transfer capacity at cathode–electrolyte contacts as decohesion accumulates. Other degradation mechanisms that may contribute to capacity fade – including solid-electrolyte-interphase (SEI) growth, interfacial (electro)chemical

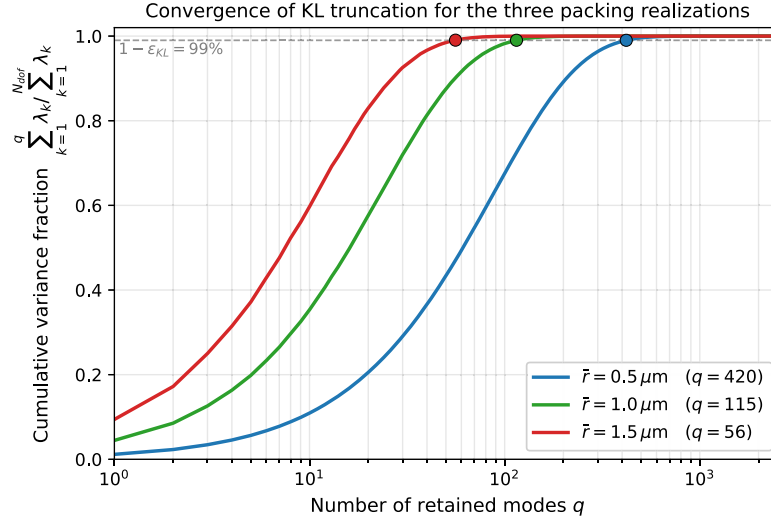
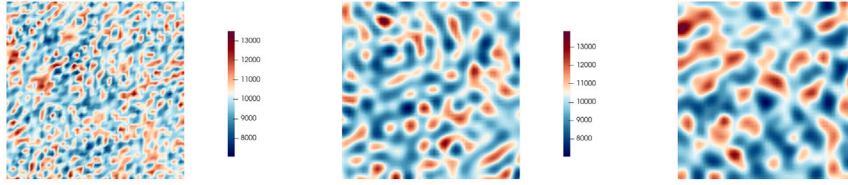


Fig. 5. Cumulative eigenvalue fraction $\sum_{k=1}^q \lambda_k / \sum_{k=1}^{N_{\text{def}}} \lambda_k$ as a function of the number of retained KL modes q , for the three packing realizations of Fig. 3. Filled markers indicate the smallest q satisfying the 1% truncation criterion of (26). Correlation lengths of the squared-exponential kernel are set equal to the mean particle diameter $2\bar{r}$ for each case.



(a) One realization of $E(\mathbf{x})$ following f_G^1 (b) One realization of $E(\mathbf{x})$ following f_G^2 (c) One realization of $E(\mathbf{x})$ following f_G^3

Fig. 6. One realization of the Young's modulus random field $E(\mathbf{x})$ for each of the three radius distributions (f_G^1 , f_G^2 , f_G^3) shown in Fig. 3. Analogous random fields are generated for the ionic conductivity and the ion diffusivity following the same procedure.

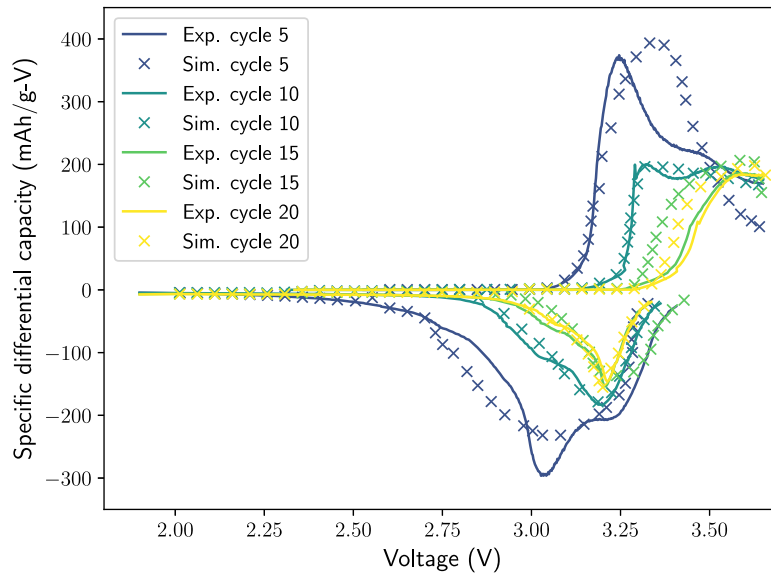


Fig. 7. Specific differential capacity of the half cell at cycle counts 5, 10, 15, and 20. Experimental data are plotted as solid lines, and simulation data points are marked as crosses. The cycle-to-cycle evolution of the simulated curves is driven entirely by the accumulation of interfacial decohesion damage D described in Section 2.3; no SEI growth, interfacial chemical reactions, or bulk cathode degradation are modeled.

Table 3
Hierarchy of characteristic length scales in the present model of the composite cathode.

Length scale	Model value	Notes
NMC811 primary crystallite	not resolved (lumped)	~ 100 nm (literature; [10])
LPSCl solid-electrolyte particle radius	$r \in \{0.5, 1.0, 1.5\} \mu\text{m}$ ($\delta = 0.1$)	three parametric cases (Section 5)
NMC811 secondary particle radius	$r = 3 \mu\text{m}$ ($\delta = 0.1$)	homogeneous continuum body
Cathode–electrolyte interface	1-D cohesive-zone interface	traction–separation law of Section 2.3
KL grid spacing h^e	$0.36 \mu\text{m}$	Fredholm eigenproblem discretization
RVE side length	$18 \mu\text{m}$	two-dimensional RVE

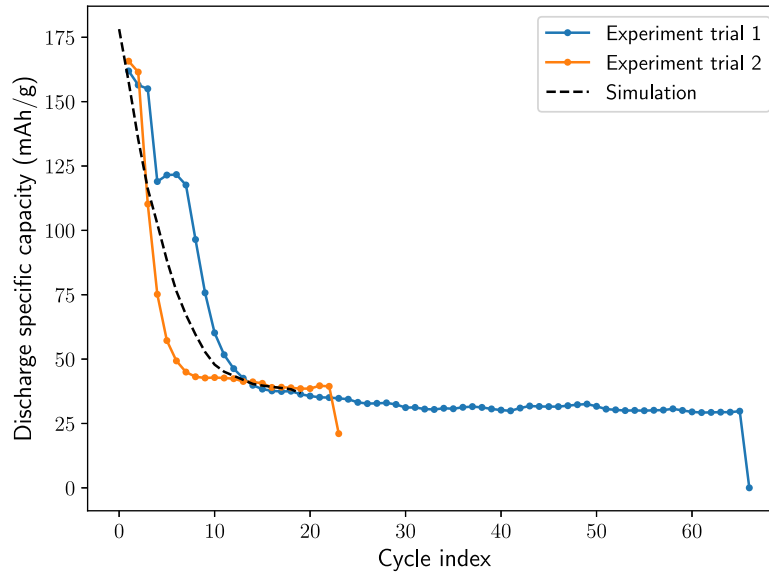


Fig. 8. Depth of discharge as a function of cycle count. Two experimental trials are shown as solid lines. The simulated depth of discharge curve is the dashed line.

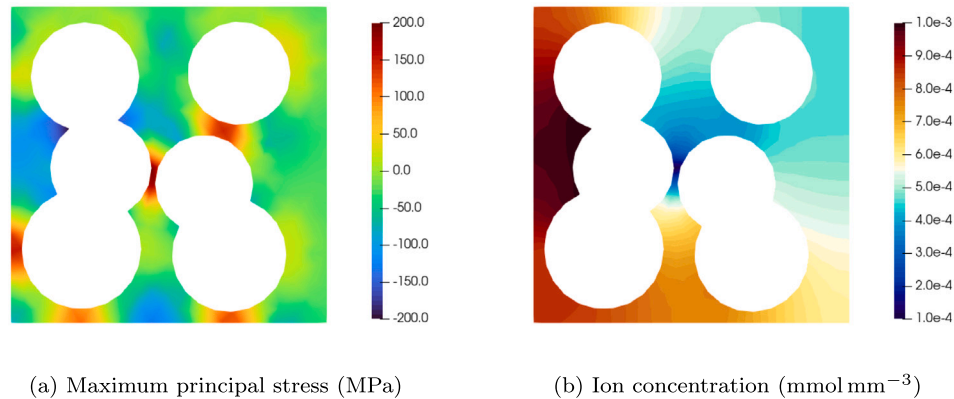


Fig. 9. Contour plots of the maximum principal stress and the ion concentration at the end of the 20th charging cycle.

reactions that consume active material, and bulk cathode degradation such as NMC phase transitions or intra-particle cracking – are not resolved in the present formulation, and the NMC811 particles themselves are treated as homogeneous elastic-swelling bodies without evolving material state; the LPSCl electrolyte is treated as an ionic conductor without an explicit interphase-growth model. SEI-like interphase growth is generally observed to be less pronounced in $\text{Li}_{0.5}\text{In}/\text{LPSCl}$ systems than in conventional liquid–electrolyte cells with Li-metal or graphite anodes, and Li-inventory loss from the $\text{Li}_{0.5}\text{In}$ reservoir is expected to be small over the 20-cycle window considered here. The fact that the model, driven by interfacial decohesion alone, reproduces the

observed evolution of the specific differential capacity and the depth of discharge (Figs. 7 and 8) is consistent with a significant mechanical-decohesion contribution to the observed capacity fade over this cycling window; a more complete attribution across the co-occurring mechanisms would require additional experiments and a multi-mechanism model, and is identified as future work in the Conclusions.

Specific differential capacity is plotted in Fig. 7 at cycle counts 5, 10, 15, and 20. Depth of discharge is shown in Fig. 8.

At cycle 20, the contour plot of the maximum principal stress and the ion concentration are shown in Fig. 9. The normal traction attains its maximum between the two center cathode particles (Fig. 9(a)). Over

the 20 cycles, a critical amount of interface damage has accumulated, effectively blocking the ion transfer for the top right quarter of the RVE (Fig. 9(b)), leading to a catastrophic degradation in the depth of discharge.

Conclusions

We have developed a stochastic continuum modeling framework to study microstructure-driven interface degradation in all-solid-state batteries. By embedding non-Gaussian, correlated random fields into a physics-based electro-chemo-mechanical model, the framework captures particle-scale heterogeneity of the solid electrolyte and its impact on stress localization and interfacial decohesion. Application to a half-cell containing NMC811 cathode particles and LPSCl electrolyte demonstrates that localized fluctuations in elasticity, ionic conductivity, and diffusivity significantly influence the evolution of interface damage, leading to early blockage of ion transport and catastrophic failure after repeated cycling. The proposed methodology bridges the gap between idealized continuum descriptions and realistic microstructural variability, offering predictive insights into degradation pathways and informing material design, interface engineering, and processing strategies. Future work will focus on incorporating multi-scale coupling with particle-resolved simulations, extending the framework to include bulk damage–transport coupling (e.g., through a phase-field crack representation that locally degrades the electrolyte's ionic conductivity), and experimental validation across diverse ASSB chemistries.

CRediT authorship contribution statement

Tianchen Hu: Writing – original draft, Validation, Software, Methodology, Formal analysis, Conceptualization. **Chen-Jui Huang:** Writing – review & editing, Validation, Investigation, Data curation. **Shirley Meng:** Writing – review & editing, Supervision, Resources, Project administration, Funding acquisition. **Bipul Barua:** Writing – review & editing, Methodology, Investigation.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

The work of TH and BB was sponsored by the U.S. Department of Energy under Contract No. DE-AC02-06CH11357 with Argonne National Laboratory, managed and operated by UChicago Argonne LLC. Funding was provided by the Joint Task Force Initiative (JTFI) at the University of Chicago (110007622).

Data availability

Data will be made available on request.

References

- [1] Joshi Aniruddha, Mishra Dillip Kumar, Singh Rajendra, Zhang Jiangfeng, Ding Yi. A comprehensive review of solid-state batteries. *Appl Energy* 2025;386:125546.
- [2] Tang Xiaojun, Feng Mengqi, Lv Wenhao, Lv Song. Applications of all-solid-state lithium-ion batteries across wide temperature ranges: Challenges, progress, and perspectives. *Adv Energy Mater* 2025;2500479.
- [3] Hong Bolong, Gao Lei, Li Changping, Lai Genming, Zhu Jinlong, Huang Dubin, Zuo Yunxing, Yin Wen, Sun Mengyu, Zhao Shusen, et al. All-solid-state batteries designed for operation under extreme cold conditions. *Nat Commun* 2025;16(1):143.
- [4] Wang Shuo, Fang Ruyi, Li Yutao, Liu Yuan, Xin Chengzhou, Richter Felix H, Nan Ce-Wen. Interfacial challenges for all-solid-state batteries based on sulfide solid electrolytes. *J Mater* 2021;7(2):209–18.
- [5] Taghikhani Kasra, Huber William, Weddle Peter J, Zaeem Mohsen Asle, Berger JR, Kee Robert J. Modeling coupled electro-chemo-mechanical phenomena within all-solid-state battery composite cathodes. *J Mech Phys Solids* 2025;198:106060.
- [6] Duan Jian, Fuchs Till, Mogwitz Boris, Minnmann Philip, Zuo Tong-Tong, Henss Anja, Janek Jürgen. Solid electrolyte cracking due to lithium filament growth and concept of mechanical reinforcement—An operando study. *Mater Today* 2023;70:33–43.
- [7] Bucci Giovanna, Swamy Tushar, Chiang Yet-Ming, Carter W Craig. Modeling of internal mechanical failure of all-solid-state batteries during electrochemical cycling, and implications for battery design. *J Mater Chem A* 2017;5(36):19422–30.
- [8] Wan Ting Hei, Ciucci Francesco. Electro-chemo-mechanical modeling of solid-state batteries. *Electrochim Acta* 2020;331:135355.
- [9] Lu Xuefeng, Gao Xu, Ren Junqiang, Li Cuixia, Guo Xin, Yan Xiaobin, La Peiqing. Effects of diverse metal adsorptions on the electronic and optical properties of the β -Si₃N₄ (2 0 0) surface: A first-principles study. *Comput Mater Sci* 2018;151:296–306.
- [10] Singh Avtar, Song Jihun, Li Wei, Martin Trevor, Xu Hongyi, Finegan Donal P, Zhu Juner. Microstructure-chemomechanics relations of polycrystalline cathodes in solid-state batteries. *Extrem Mech Lett* 2024;69:102164.
- [11] Wu Jian-Ying. A generalized phase-field cohesive zone model (μ PF-CZM) for fracture. *J Mech Phys Solids* 2024;192:105841.
- [12] Liu Sijia, Wang Yunteng, Peng Chong, Wu Wei. A thermodynamically consistent phase field model for mixed-mode fracture in rock-like materials. *Comput Methods Appl Mech Engrg* 2022;392:114642.
- [13] Huang Chen-Jui, Oh Jin An Sam, Vicencio Marta, Hu Tianchen, Yang Hedi, Burrow James N, Song Yen-Fang, Yin Gung-Chian, Shevchenko Pavel, Wiaderek Kamila M, et al. X-ray micro-computed tomography for structural analysis of all-solid-state battery at pouch cell level. *ACS Energy Lett* 2025;10(7):3459–70.
- [14] Fu Jinlong, Tan Wei. Stochastic reconstruction of multiphase composite microstructures using statistics-encoded neural network for poro/micro-mechanical modelling. *Comput Methods Appl Mech Engrg* 2025;441:117986.
- [15] Hu Tianchen, Messner Mark C, Barai Pallab, Barua Bipul. A three-dimensional, thermodynamically and variationally consistent, fully coupled, electro-chemo-thermo-mechanical model of solid-state batteries. *J Electrochem Soc* 2023;170(12):123501.
- [16] Camanho Pedro P, Dávila Carlos G. Mixed-Mode Decohesion Finite Elements for the Simulation of Delamination in Composite Materials. Technical report, 2002.
- [17] Grigoriu Mircea. Existence and construction of translation models for stationary non-Gaussian processes. *Probabilistic Eng Mech* 2009;24:545–51.
- [18] Jaynes ET. Information Theory and statistical mechanics I. *Phys Rev* 1957;106(4):620–30.
- [19] Jaynes ET. Information Theory and statistical mechanics II. *Phys Rev* 1957;108(2):171–90.
- [20] Ghanem RG, Spanos PD. Stochastic Finite Elements: A Spectral Approach (Revised Edition). Dover; 2003.
- [21] Le Maitre O, Knio O. Spectral methods for uncertainty quantification. Springer Netherlands; 2010.
- [22] Hu Tianchen, Messner Mark C, Barai Pallab, Barua Bipul. EEL: A MOOSE-based Application for 3D Electro-Chemo-Thermo-Mechanical Modeling of Solid-State Batteries. Technical Report ANL-22/67, Argonne, IL, USA: Argonne National Laboratory; 2022.
- [23] Permann Cody J, Gaston Derek R, Andrš David, Carlsen Robert W, Kong Fande, Lindsay Alexander D, Miller Jason M, Peterson John W, Slaughter Andrew E, Stogner Roy H, Martineau Richard C. MOOSE: Enabling massively parallel multiphysics simulation. *SoftwareX* 2020;11:100430.
- [24] Balay Satish, Abhyankar Shrirang, Adams Mark F, Benson Steven, Brown Jed, Brune Peter, Buschelman Kris, Constantinescu Emil, Dalcin Lisandro, Dener Alp, et al. PETSc web page. 2024. <https://petsc.org/>.
- [25] Hernandez Vicente, Roman Jose E, Vidal Vicente. SLEPC: A scalable and flexible toolkit for the solution of eigenvalue problems. *ACM Trans Math Softw (TOMS)* 2005;31(3):351–62.
- [26] Wang Weixin, Wang Hui, Dai Guozhong, Wang Hongan. Visualization of large hierarchical data by circle packing. In: Proceedings of the SIGCHI conference on human factors in computing systems. CHI '06, New York, NY, USA: Association for Computing Machinery; 2006, p. 517–20. <http://dx.doi.org/10.1145/1124772.1124851>.