Mathematical modelling for all-solid-state battery: (se|sse)-Interface

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Mathematical modelling for the next-generation All-solid-state batteries: Nucleation (SE|SSE)^(*)-interface

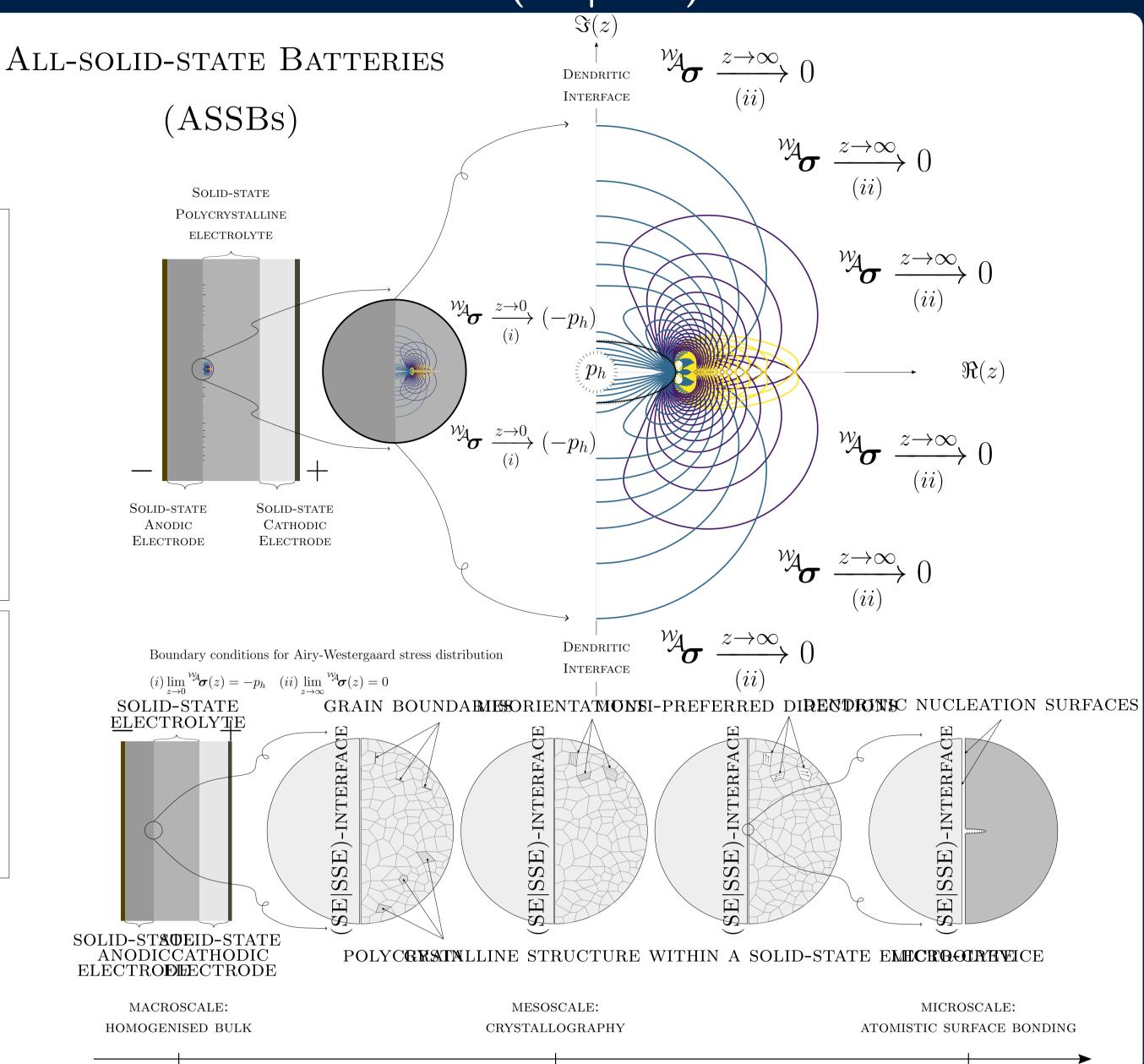
Rechargeable Lithium-ion battery (LIB) is at the heart of every electric vehicle (EV), portable electronic device, and energy storage system [1]. Nowadays, LIBs enable human life more efficient and help to solve global environment issues thanks to EVs' zero However, conventional LIB (c-LIB) is emission. sensible to temperature and pressure, hence, flammable and explosive, which is undesirable. This bottleneck is | causing crevice, degradation of ionic conductivity, and mainly due to liquid-based electrolyte found in c-LIBs.

All-solid-state battery (ASSB) is one of promising candidates to overcome bottlenecks of c-LIBs. Thanks to solid-state electrolyte (SSE), ASSB is highly stable towards temperature and pressure. Nevertheless, Limetal dendrite triggered at (SE|SSE)-interface [5] is the main drawback of ASSB since these dendritic threads extrapolate into SSE grain boundary network, the probability of short-circuit, which is unfavorable.

Next-generation All-solid-state battery (ng-ASSB) with a consideration of nucleation criterion defined by

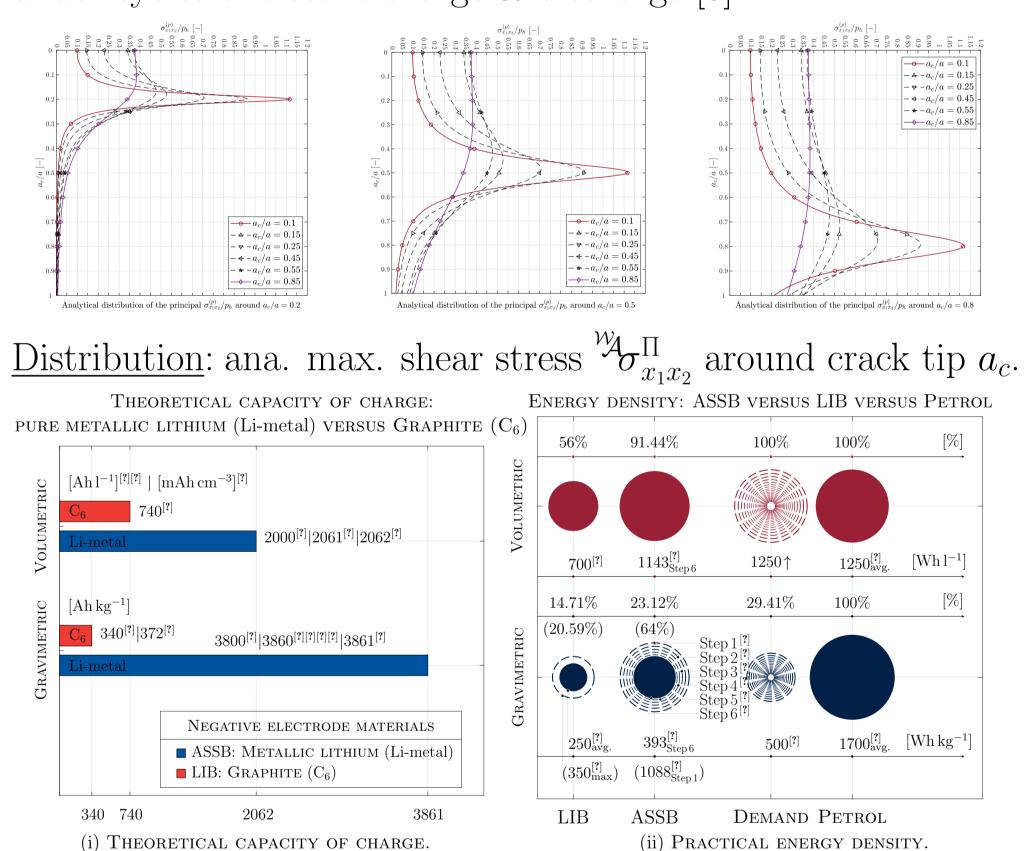
$$a_{\text{Griffith}} := a^* = \arg\min_{a \in \mathbb{R}} \iiint_{\Omega} f(a, \boldsymbol{u}, \boldsymbol{\theta}; \lambda, \mu, \boldsymbol{d}^{(\star)} \otimes \boldsymbol{d}^{(\star)}) \, d\Omega - \iint_{\Gamma} f(a; \gamma) \, d\Gamma \bigg|_{\boldsymbol{d}}$$

where \boldsymbol{u} displacement field, θ temperature field, a crevice length, λ, μ Lamé constants, $\boldsymbol{d}^{(\star)} \otimes \boldsymbol{d}^{(\star)}$ embedded misorientation structural tensor, and γ cracking-surface energy density, can help to improve ASSB performance.



(SE|SSE)-Interface Analysis

Interface between solid electrode and solid-state electrolyte (SE|SSE) taking place at space charge layer (SCL) [2] found in ASSBs critically exhibits mechanical and electrochemical instability [3]. This evidence points directly to the fact that the soft metallic li anode is erroneously prone to triggering dendrites, under cycles of electric charge & discharge [5].



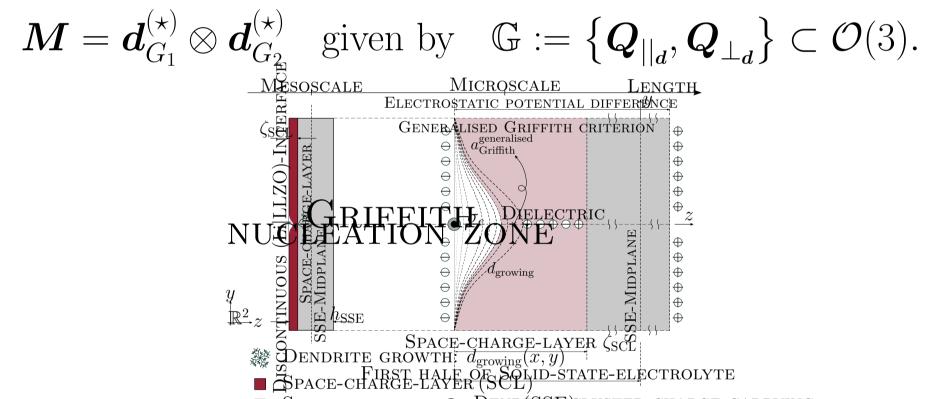
Next-generation All-solid-state battery

Nucleation criterion governs the instable (SE|SSE)-interface [3] Discharging process: Li-ion Stripping $\blacksquare \rightarrow \blacksquare$ METAL-OXIDE SOLID-STATE NEGATIVE ELECTRODE Li-ion Transpor Discharge direction -)-ELECTRODE+)-ELECTRODE RIFFITH NUCLEATION ZONE DISCONTINUOUS (Li|LLZO)-INTERSACE

Thermodynamic consistency is satisfied, followed by [2]. Closure Ω is fulfilled by 15 moments, followed by [4].

Embedded structural-tensor in SSE

Polycrystalline garnet-type SSE [5] such as LLZO exhibit grain boundary network, and grains with variation of {size, shape} under microscopic observation. Hence, this microstructure is potentially prone to nuances of destruction.



Consequentially, dendrites contribute to degradation of ionic conductivity and tiny-cracks tracing along grain boundaries.

Nucleation interface: Taking place at the critically dendritic (SE|SSE)-interface

Coupled fields: Displacement field u and temperature field θ ; structural tensor M

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Governing conservation equations

$$\frac{d}{dt} \int_{\Omega} (\cdot) \ d\Omega = \int_{\Omega} (\cdot)^{\text{action}} \ d\Omega + \int_{\partial \Omega} (\cdot)^{\text{action}} \ d\partial\Omega + \int_{\Omega} (\cdot)^{\text{production (+/-)}} \ d\Omega$$

used to describe balance of mass, conservation of linear momentum, conservation of angular momentum, and conservation of energy with $\rho(\boldsymbol{x},t)$ is mass density per unit volume (puv); $\boldsymbol{b}(\boldsymbol{x},t)$ body force puv; $\boldsymbol{v}(\boldsymbol{x},t)$ velocity; $e(\boldsymbol{x},t)$ internal energy puv; q(x,t) heat flux; r(x,t) heat source puv; σ Cauchy stress and ε infinitesimal strain. Then, the governing partial differential equation (PDE) for the thermodynamically consistent coupled problem with deformation field \boldsymbol{u} as its variable takes the form

$$\partial_t oldsymbol{u} +
abla \cdot \left(\overset{4}{\mathbb{C}}^{f_{ ext{alocation}}(\lambda, \mu, oldsymbol{d}_{G_i, i=1,...,N}^R, oldsymbol{d}^E; oldsymbol{x})} :
abla oldsymbol{u}^{(s)}
ight) +
ho oldsymbol{b} = -
ho
abla V_e,$$

where $V_e: \mathbb{R}^3 \to \mathbb{R}$ is the electric potential applied globally on ASSB. Due to nature setting of ASSB taking the form (SE|SSE|SE) the electric potential becomes uniform.

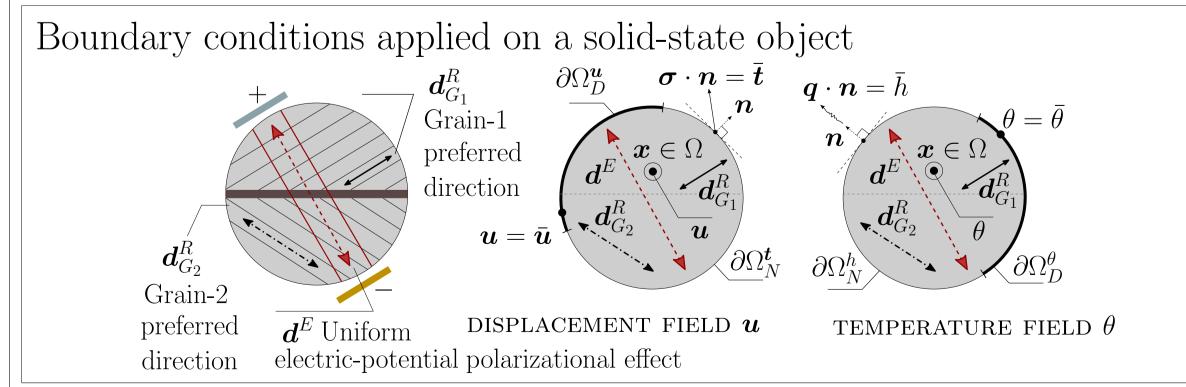
Strain **energy** is based on the deformation of SSE due to dendrite formation at (SE|SSE)-interface

 $\iiint_{\Omega} f(a, \boldsymbol{u}; \lambda, \mu, \boldsymbol{d} \otimes \boldsymbol{d}) d\Omega$

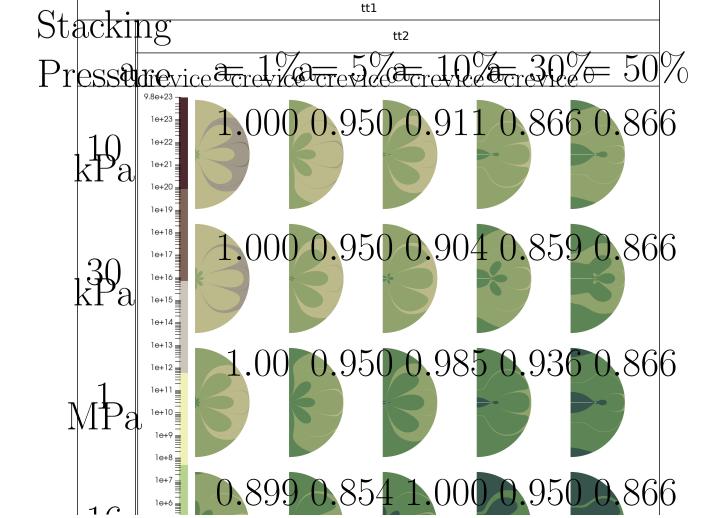
Surface energy is analysized based on the open crevice cracking at (SE|SSE)interface affected by prescribed pressure

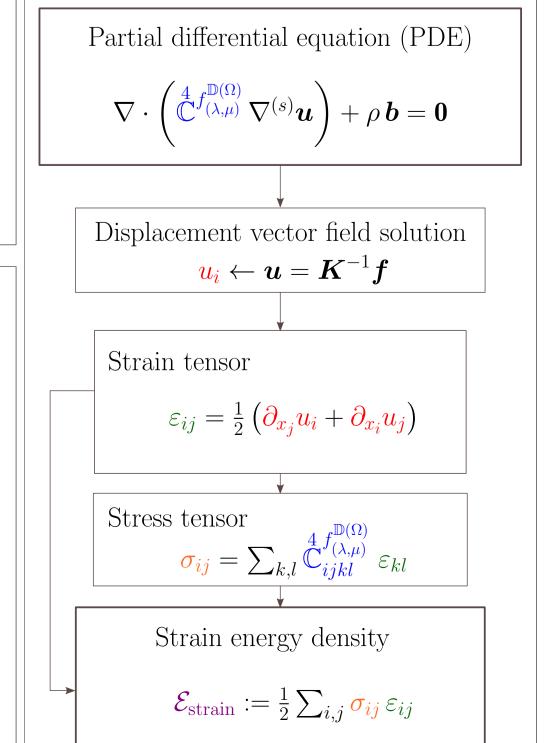
$$\iint_{\Gamma} f(a;\gamma) \, d\Gamma$$

Analysis: Airy-Westergaard fcn. used for stress analysis: (i) max. shear stress and (ii) principal stresses $| \mathcal{V}_{\mathcal{A}} : \begin{cases} \mathbb{C} \to \mathbb{C}, \\ z \mapsto \mathcal{V}_{\mathcal{A}}(z) := \Re(\oint_{\mathbb{C}} \mathcal{K}^{(\star)} dz) + x_2 \Im(\oint_{\mathbb{C}} \mathcal{K}^{(\star)} dz), \end{cases} \mathcal{K}^{(\star)} : \begin{cases} \mathbb{C} \to \mathbb{C}, \\ z \mapsto \mathcal{K}^{(\star)} := -p_h + p_h/\sqrt{1 - a^2/z^2}, \end{cases}$ where a the crevice length, p_h pressure at the opening crevice on dendritic interface, and $\forall \{p_h, a\} \in \mathbb{R}_+$.



Numerical spectral of *Griffith* criterion at (SE|SSE) yields (xx-yy-xy)





Strain solution takes the following form

 $\frac{1}{2} \sum_{\alpha=1}^{\mathcal{N}_{\text{node}}^{\Omega^e}} \left(\sum_{L=1}^{\mathcal{N}_{\text{dof}}^{\Omega^{\text{node}}}} N_{,\xi_L}^{\alpha} \xi_{L,x_k} \boldsymbol{u}_k^{\alpha} + \sum_{K=1}^{\mathcal{N}_{\text{dof}}^{\Omega^{\text{node}}}} N_{,\xi_K}^{\alpha} \xi_{K,x_l} \boldsymbol{u}_l^{\alpha} \right)$

FEM: Strain energy density