

MATHEMATICAL MODELLING FOR ALL-SOLID-STATE BATTERY

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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 [5]. Nowadays, LIBs enable human life more efficient and help to solve global environment issues thanks to EVs' zero emission. However, conventional LIB (c-LIB) is sensible to temperature and pressure, hence, flammable and explosive, which is undesirable. This bottleneck is 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, Li-metal dendrite triggered at (SE|SSE)-Interface is the main drawback of ASSB since these dendritic threads extrapolate into SSE grain boundary network, causing crevice, degradation of ionic conductivity, and the probability of short-circuit, which is unfavorable [10].

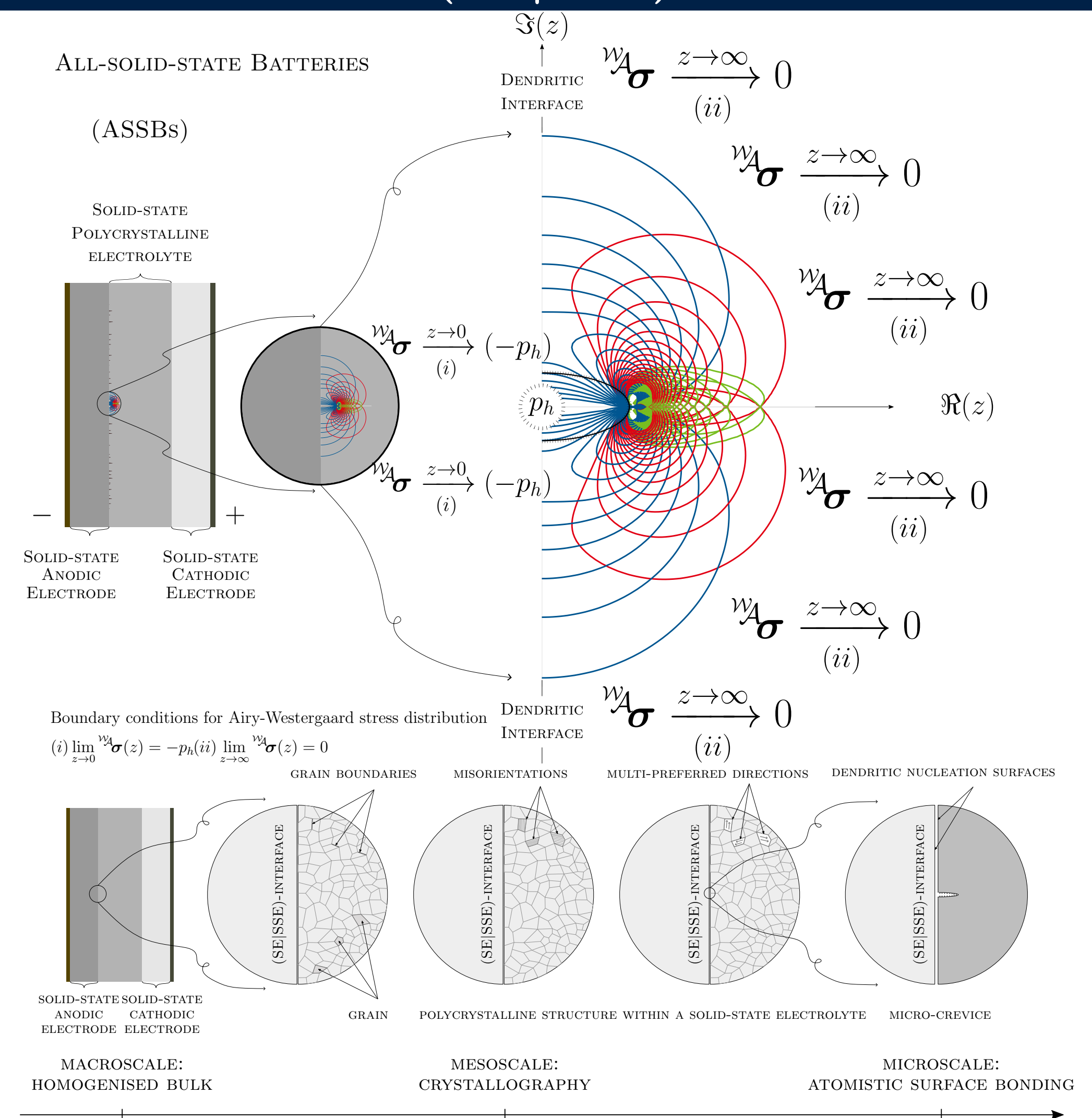
Next-generation All-solid-state battery (ng-ASSB) with a consideration of **nucleation criterion** defined by

$$\rho_{\text{SCL}} \frac{D^2 \mathbf{u}_{\text{SCL}}}{Dt^2} + \nabla \cdot (\mathbb{C}(\lambda, \mu) : \nabla \mathbf{u}_{\text{SCL}}^{(s)}) + \rho_{\text{SCL}} \mathbf{b} = -\rho_{\text{SCL}} V_e, \quad (1)$$

$$\text{s.t. } a_{\text{Griffith}}^{\text{generalised}} := a^* = \arg\left\{\min_{a \in \mathcal{V}} \iint_{\Omega} f(a_{\text{crevice}}, \mathbf{u}_{\text{SCL}}, \theta_{\text{SCL}}, n^{\text{Li}^+}; \lambda, \mu, \mathbf{d}_{\text{SCL}} \otimes \mathbf{d}_{\text{SCL}}) d\Omega - \iint_{\Gamma} f(a_{\text{crevice}}, \gamma_s) d\Gamma\right\}, \quad (2)$$

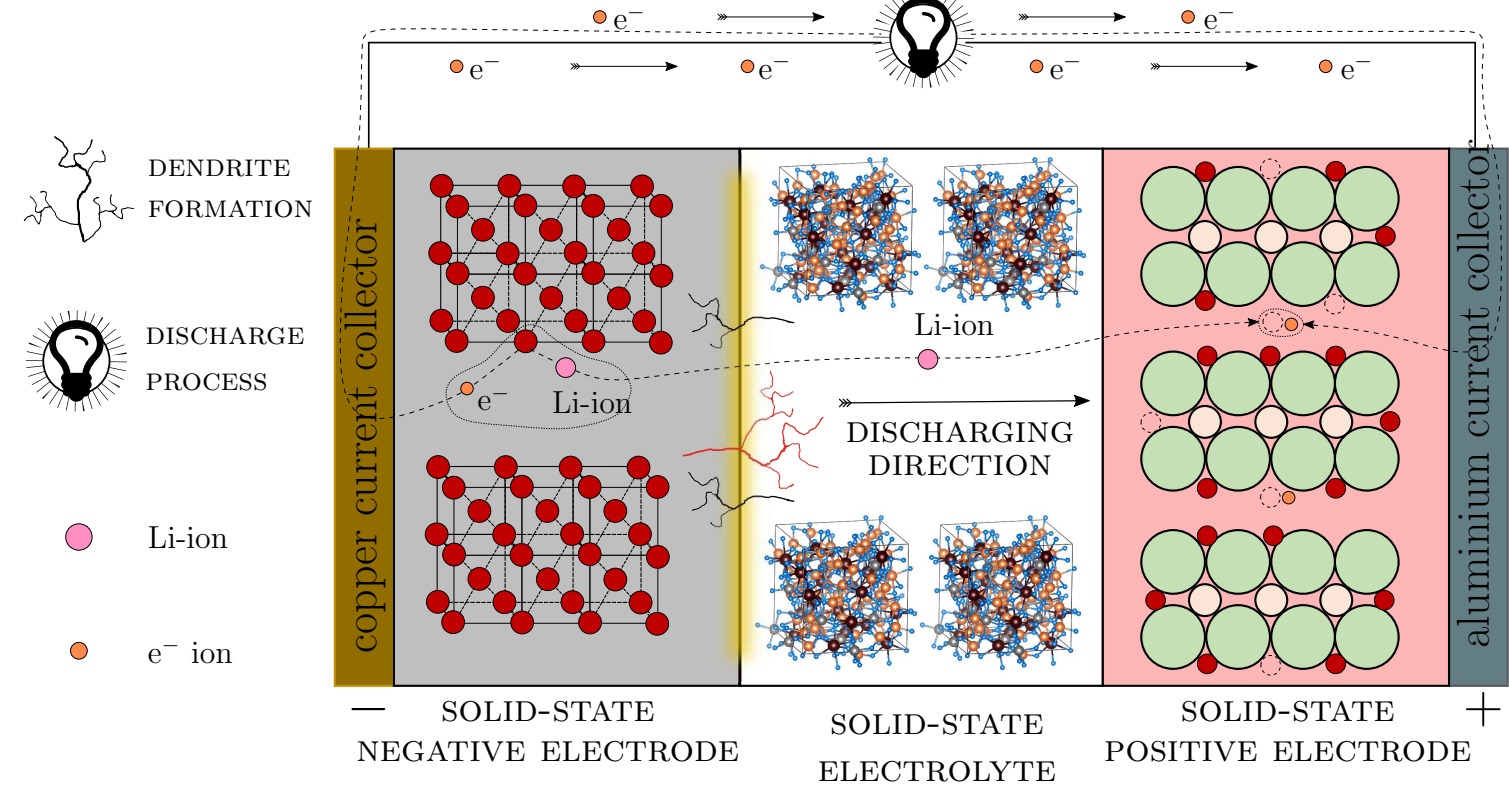
hold for $\forall a \in \mathcal{V}$. Here, $V_e : \mathbb{R}^3 \rightarrow \mathbb{R}$ is the electric potential applied globally on ASSB. Due to nature setting of ASSB taking the form (SE|SSE|SE) the electric potential becomes uniform. Additionally, \mathbf{u} is the displacement field, θ temperature field, a crevice length, λ, μ Lamé constants, $\mathbf{d} \otimes \mathbf{d}$ embedded misorientation SCL structural tensor, and γ_s cracking-surface energy density, can help to improve ASSB performance [1][2].

Aim: The study is with the purpose of gaining a better insight into dendrite nucleation and formation in ASSB.



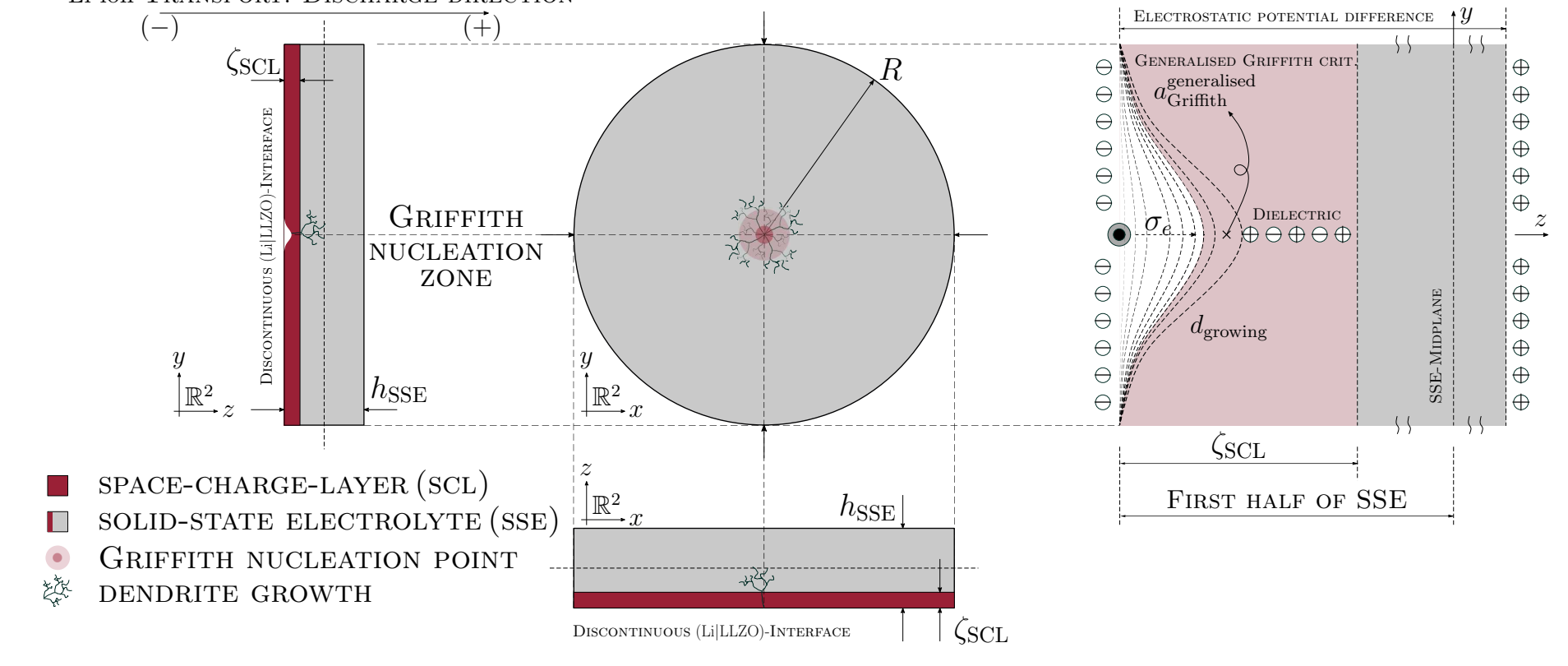
Next-generation All-solid-state battery

Griffith nucleation criterion governs (SE|SSE)-Interface [4].



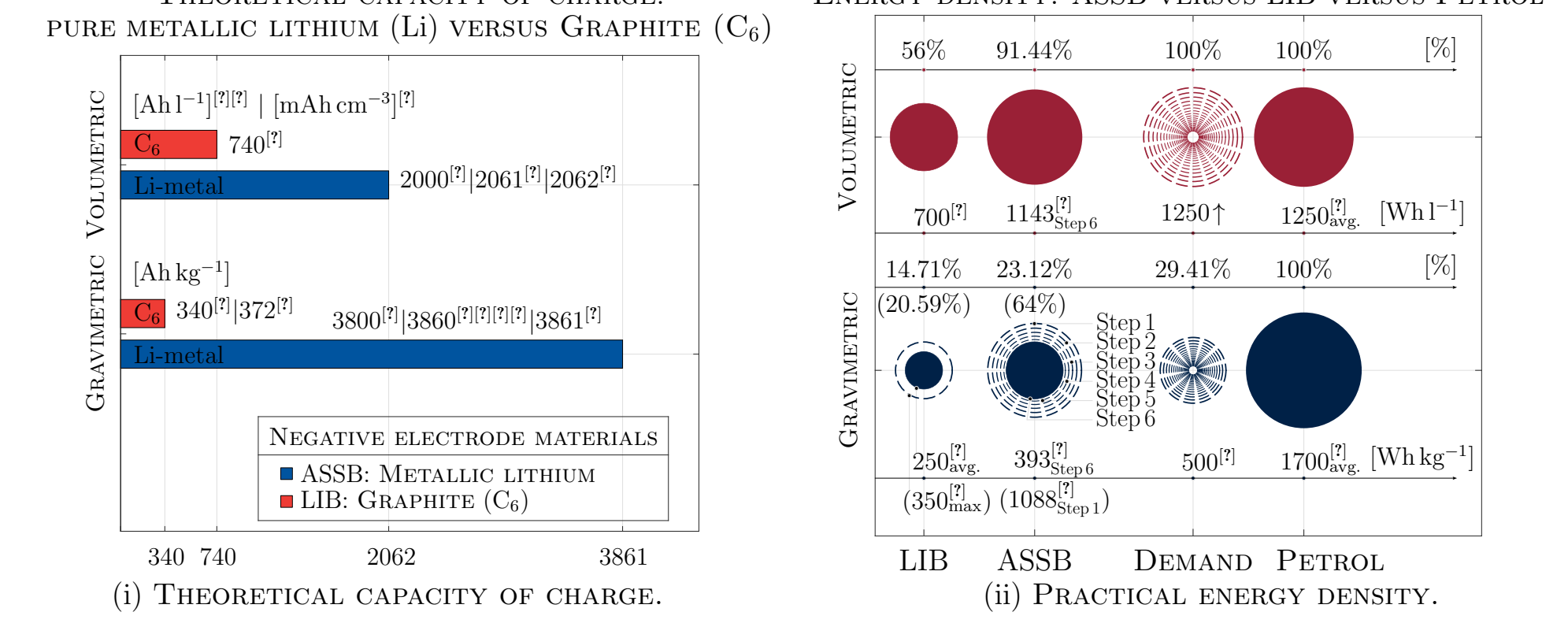
Observation: Space-charge Layer

SCL manifests in ASSB [8], predictably in Semiconductors.



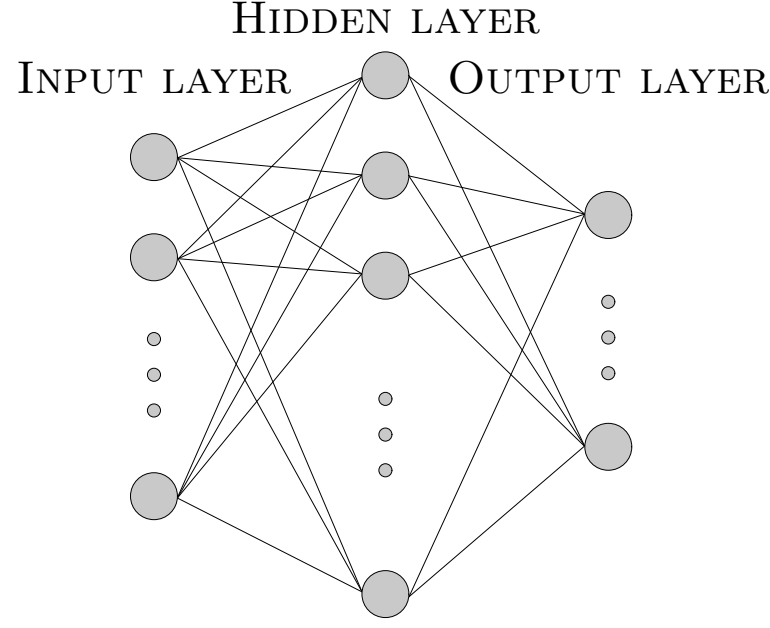
Motivation: Energy density landscape

ASSB enables **energy demand** due to (i), and followed by (ii).



Artificial Neural Networks

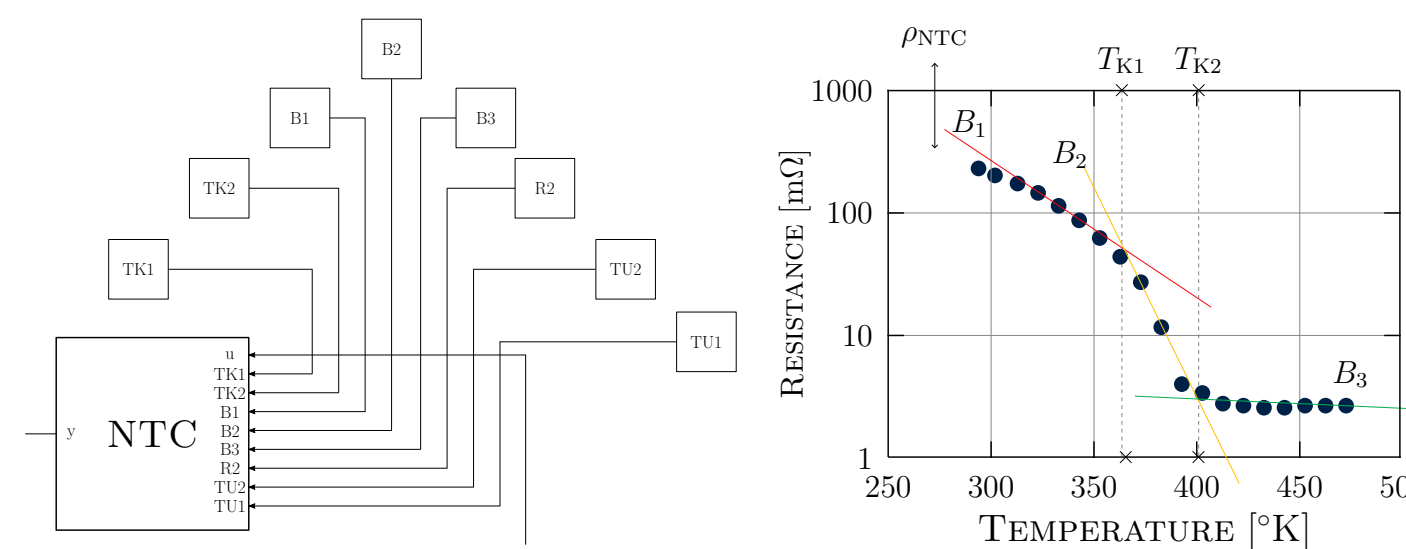
Application: Steel's property prediction.



The ANNs scheme enhances bainitic trafo. temperature prediction, validated by [9].

Semiconductor

Application: Start/Stop-System in Starter.
Use-case: BMW B47 (-25°C, 0°C, 120°C).
Optimisation: Pareto @BoschForschung. (Multi-objective optimisation framework)

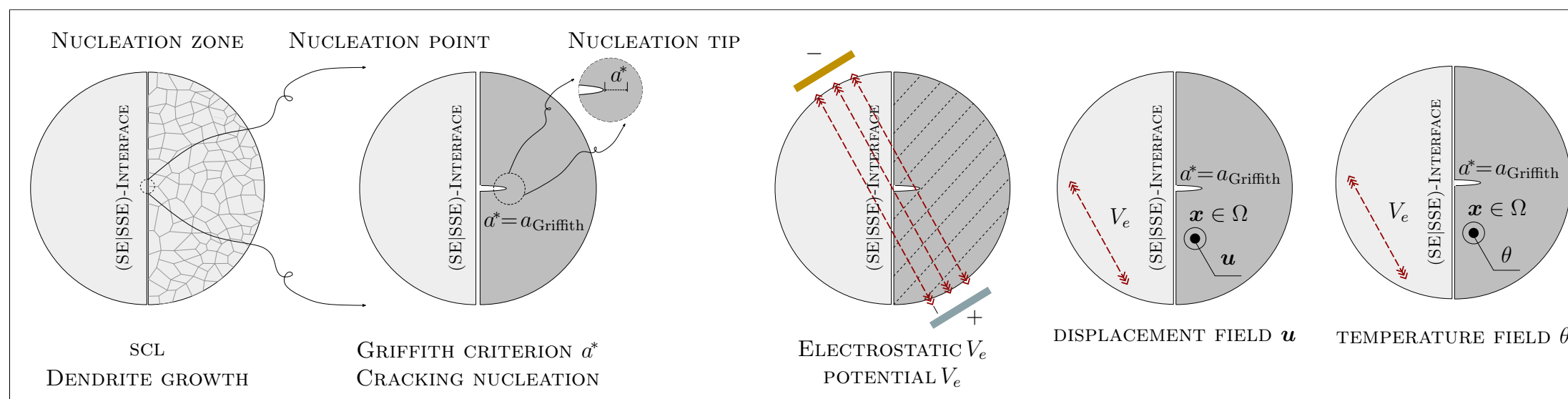


Nd/Gd Negative-Temperature Coefficient (NTC) semiconductor model validated [7].

Lithium-ion battery

Modelling: Swelling phenomena @FEM [5].
Use-case: Bosch-48-V-Battery.

State-of-the-art: Nucleation interface taking place at the unstable (SE|SSE)-Discontinuity



Coupled fields are Displacement field \mathbf{u} and Temperature field θ :

$$\mathbf{u} : \begin{cases} \Omega \times \mathbb{R}_+ \rightarrow \mathbb{R}^3, \\ (\mathbf{x}, t) \mapsto \mathbf{u}(\mathbf{x}, t), \end{cases} \quad \theta : \begin{cases} \Omega \times \mathbb{R}_+ \rightarrow \mathbb{R}, \\ (\mathbf{x}, t) \mapsto \theta(\mathbf{x}, t). \end{cases}$$

Governing conservation equations account for mass balance, linear and angular momentum, and energy conservation. These equations include variables such as mass density $\rho(\mathbf{x}, t)$, body force $\mathbf{b}(\mathbf{x}, t)$, velocity $\mathbf{v}(\mathbf{x}, t)$, internal energy $e(\mathbf{x}, t)$, heat flux $\mathbf{q}(\mathbf{x}, t)$, heat source $r(\mathbf{x}, t)$, Cauchy stress $\boldsymbol{\sigma}$, and infinitesimal strain $\boldsymbol{\varepsilon}$ per unit volume.

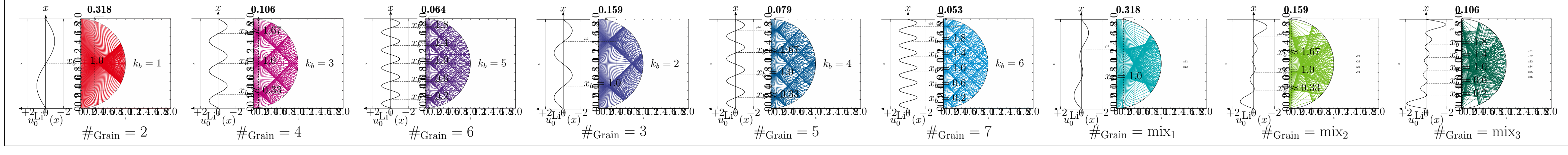
Strain energy (E_{st}) is derived from the SSE deformation due to dendrite formation at the unstable (SE|SSE)-Interface:

$$E_{\text{st}} := \iiint_{\Omega} f(a, \mathbf{u}; \lambda, \mu, \mathbf{d} \otimes \mathbf{d}) d\Omega$$

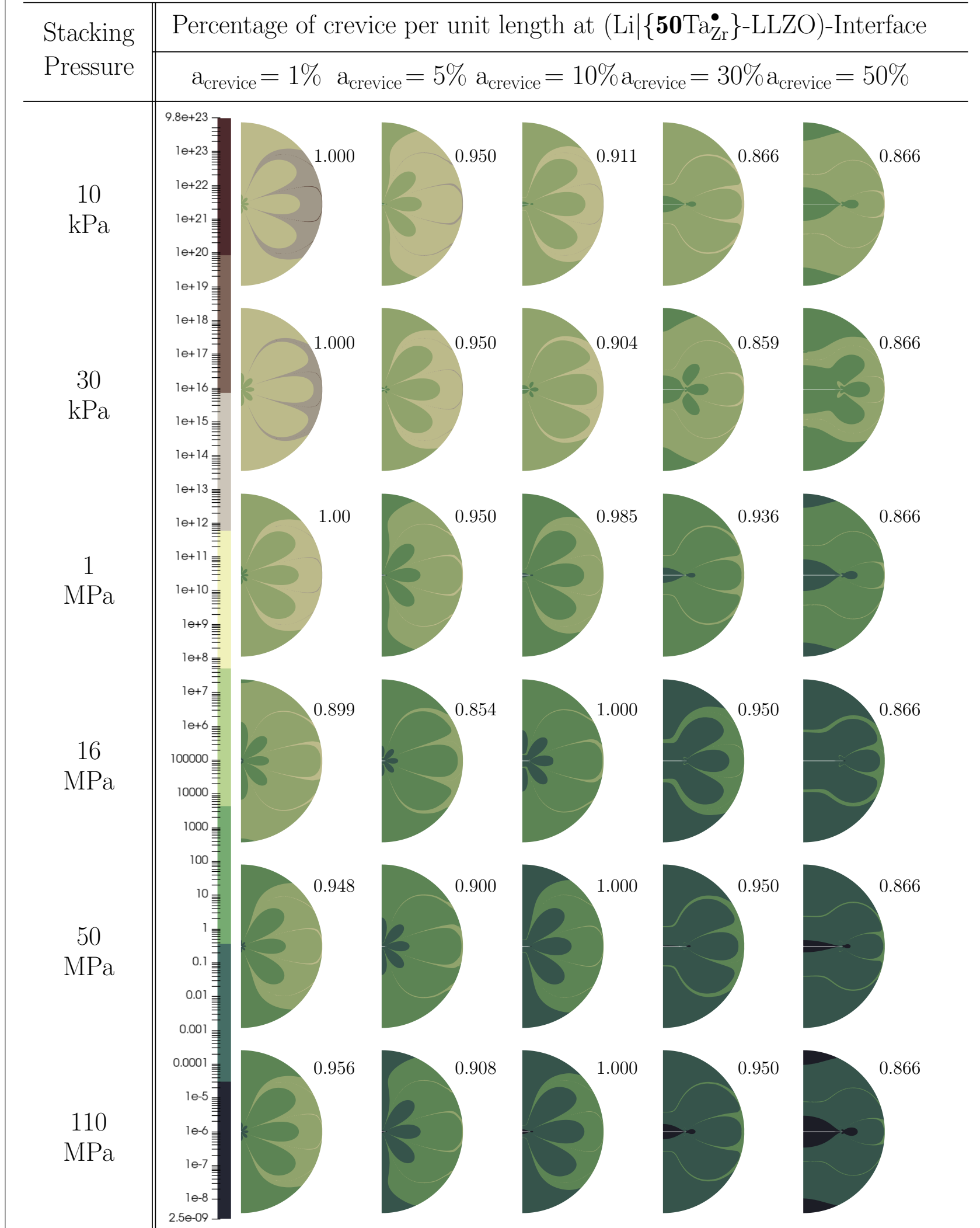
Surface energy (E_{sf}) is assessed through the analysis of crevices at the (SE|SSE)-Interface under specific pressure conditions:

$$E_{\text{sf}} := \iint_{\Gamma} f(a; \gamma) d\Gamma$$

Shock: Li-ion transport leads to shocking phenomena at the discontinuous (SE|SSE)-Interface, influenced by the distribution and size of grains.



Griffith criterion on (100-Li-termination)-{50Ta₂}-doped-Li₇La₃Zr₂O₁₂ at T=298°K



Solution:

Analysing system of equations (1) and (2) yields a simplified Griffith nucleation criterion, incorporating Cauchy stress σ , Young's modulus E_Y , and experimentally-derived surface energy γ_s , with σ calculated using FVM based on the OpenFOAM library.

$$a_{\text{Griffith}}^{\text{simplified}} = \sqrt{\frac{2 E_Y \gamma_s}{\pi \sigma}}$$

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References

- [1] T. Vo, C. Hüter, S. Braun, M. Torrilhon, *Math. modelling for ASSB: (SE|SSE)-Interface*. Poster, Oxford Batt. Modelling Symp., University of Oxford, OBMS23, P. 66, **2023**.
- [2] T. Vo, C. Hüter, S. Braun, M. Torrilhon, *Next-gen. All-solid-state Battery (#ASSB)*. Poster, SIAM Computational Science and Eng. Conference, CSE23, Amsterdam, **2023**.
- [3] T. Vo, *Math. modelling for ASSB*. Presentation, Numerical Modelling in Applied Physics with OpenFOAM Summer School, NUMAP-FOAM, University of Cambridge, **2022**.
- [4] T. Vo, C. Hüter, S. Braun, R. Spatschek, *Mathematical modelling for All-solid-state battery: Griffith criterion*. Doctoral presentation, IEK-2, Forschungszentrum Jülich, **2020**.
- [5] T. Vo, *Modeling the swelling phenomena of li-ion batt. cells based on a numerical chemo-mech. coupled approach*. Master thesis, Robert Bosch Battery Systems GmbH, **2018**.
- [6] T. Vo, A. Gallet-Segarra, C. Bertsch, *Integration of Modelica powertrain models into Hardware in the loop env.*. PreMaster, Robert Bosch GmbH, FEBER, EAM-P2695, **2015**.
- [7] T. Vo, *Simulation environment for NTC-based voltage drop reduction in Start/Stop appl. and its optimization*. Semiconductor. Bachelor thesis, Robert Bosch GmbH, **2014**.
- [8] S. Braun, C. Yada, A. Latz, *Thermodynamically consistent model for Space-Charge-Layer formation in a solid electrolyte*. J. Physical Chemistry C, 119, 22281-22288, **2015**.
- [9] C. Hüter, X. Yin, T. Vo, S. Braun, *A pragmatic dataset augmentation approach for transformation temp. prediction in steels*. Comp. Mat. Science, Vol. 176, 109488, **2020**.
- [10] C. Hüter, S. Fu, M. Finsterbusch, E. Figgemeier, L. Wells, and R. Spatschek, *Electrode-electrolyte interface stability in SSE system*. AIMS Materials Science, 4(4):867-877, **2017**.

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