Mathematical modelling for All-solid-state battery

Tuan Vo[a,b †], Claas Hüter[b], Stefanie Braun[a]

[a]Department of Mathematics, Applied and Computational Mathematics (ACoM), RWTH Aachen University, Schinkelstraße 02, 52062 Aachen, Germany
[b]Institute of Energy and Climate Research (IEK-2), Forschungszentrum Jülich, Wilhelm-Johnen-Straße, 52428 Jülich, Germany

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].

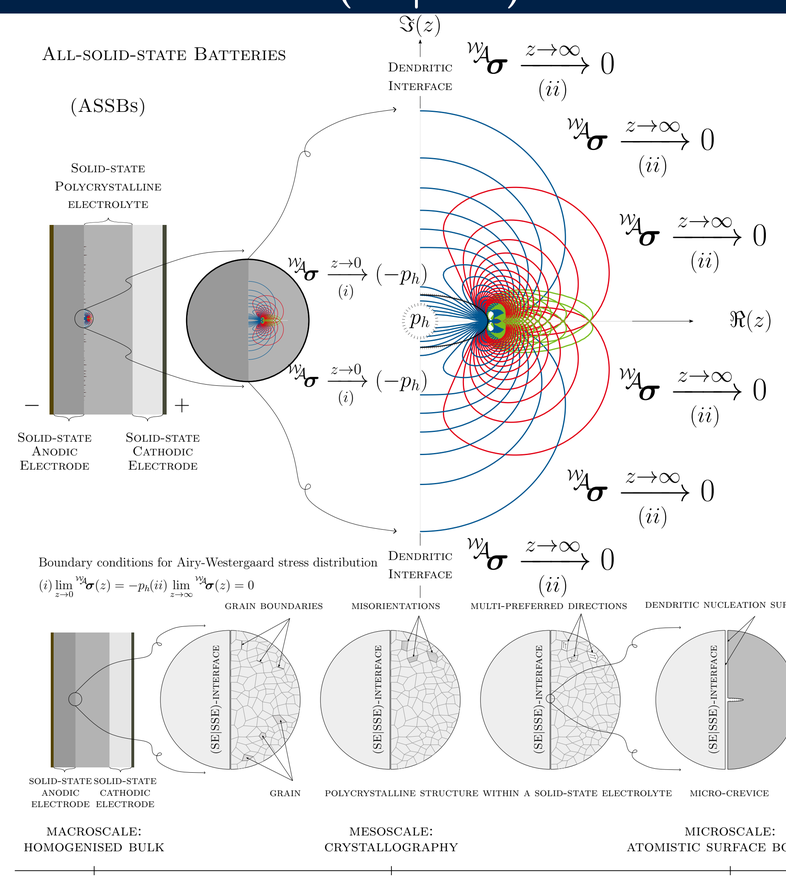
Next-generation All-solid-state battery (ng-ASSB) with a consideration of nucleation criterion defined by

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

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

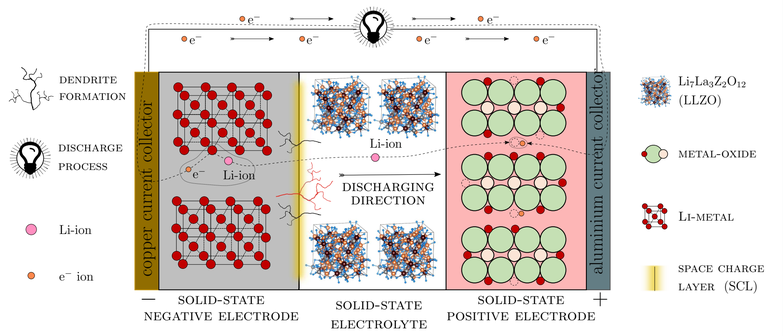
hold for $\forall\, a\in\mathcal{V}$. Here, $V_e:\mathbb{R}^3\to\mathbb{R}$ is the electric potential applied globally on ASSB. Due to nature setting of ASSB taking the form (SE|SSE|SE) the electric potential becomes uniform. Additionally, $\boldsymbol{u}$ is the displacement field, θ temperature field, a crevice length, λ,μ Lamé constants, $\boldsymbol{d}\otimes\boldsymbol{d}$ embedded misorientation SCL structural tensor, and γ 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.



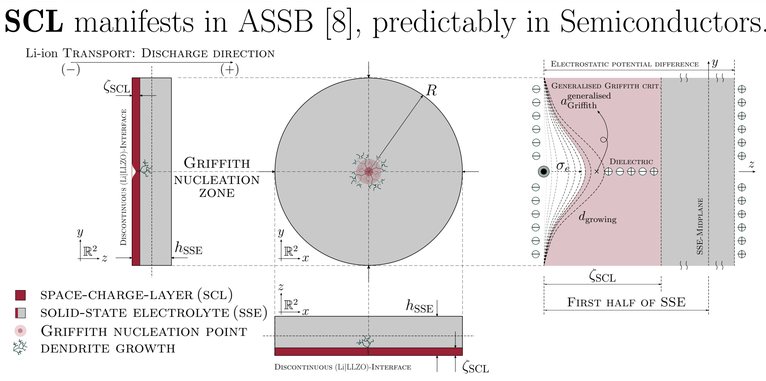
Next-generation All-solid-state battery

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



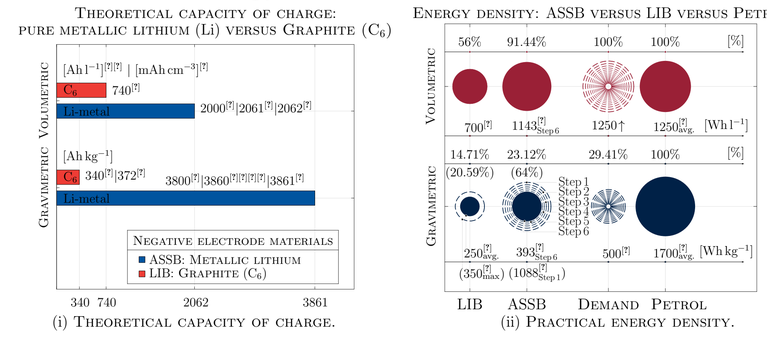
Observation: Space-charge Layer

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



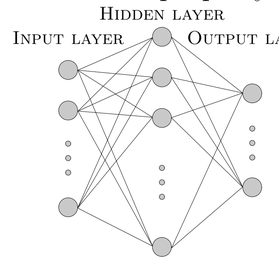
Motivation: Energy density landscape

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



Artificial Neural Networks

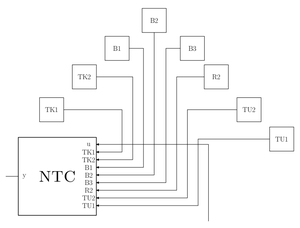
Application: Steel's property prediction.

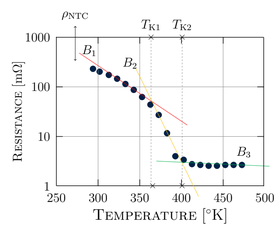


The ANNs scheme enhances bainitic trafo. temperature prediction, validated by [9].

Semiconductor

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



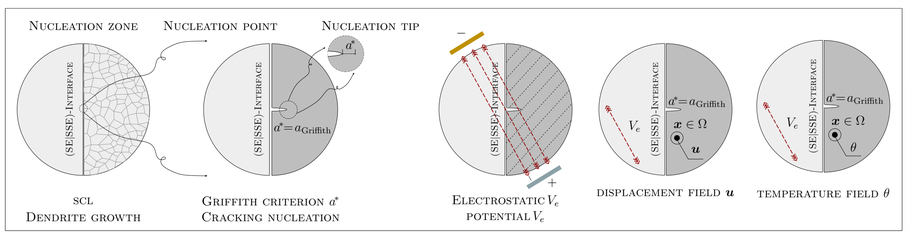


Nd/Gd Negative-Temperature Coefficient (NTC) semiconductor model validated [7].

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 $\boldsymbol{u}$ and Temperature field θ:

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

Governing conservation equations account for mass balance, linear and angular momentum, and energy conservation. These equations include variables such as mass density $\rho(\boldsymbol{x},t)$, body force $\boldsymbol{b}(\boldsymbol{x},t)$, velocity $\boldsymbol{v}(\boldsymbol{x},t)$, internal energy $e(\boldsymbol{x},t)$, heat flux $\boldsymbol{q}(\boldsymbol{x},t)$, heat source $r(\boldsymbol{x},t)$, 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:

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

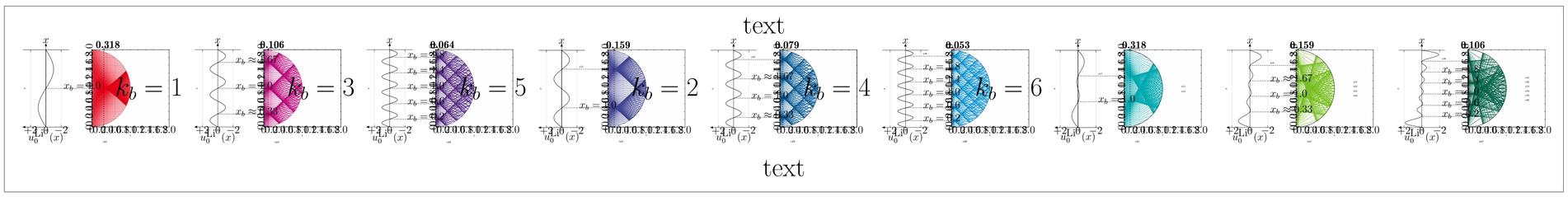
Surface energy (E_{sf}) is assessed through the analysis of crevices at the (SE|SSE)-Interface under specific pressure conditions:

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

Griffith nucleation criterion is computed based on the relation where the Cauchy stress σ is computed based on OpenFOAM library.

Griffith criterion on (**100**-Li-termination)-{**50**$Ta^{\bullet}_{Zr}$}-doped-$Li_7La_3Zr_2O_{12}$ at T=298°K
Pressure vs Standing of pre-existing crevice per unit length at (Li|{**50**$Ta^{\bullet}_{Zr}$}-LLZO)-Interface

Pressure	$a_{\mathrm{crevice}}=1\%$	$a_{\mathrm{crevice}}=5\%$	$a_{\mathrm{crevice}}=10\%$	$a_{\mathrm{crevice}}=30\%$	$a_{\mathrm{crevice}}=50\%$
10 kPa	1.000	0.950	0.911	0.866	0.866
30 kPa	1.000	0.950	0.904	0.859	0.866
1 MPa	1.00	0.950	0.985	0.936	0.866
16 MPa	0.899	0.854	1.000	0.950	0.866
50 MPa	0.948	0.900	1.000	0.950	0.866
110 MPa	0.956	0.908	1.000	0.950	0.866



Contact

Tuan Vo
vo@acom.rwth-aachen.de

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**.

Acknowledgments

T. Vo expresses sincere gratitude to the JARA-CSD research project, led by Dr. S. Braun and Dr. C. Hüter, for their crucial support. Special thanks to Prof. Dr. R. Spatschek for hosting at IEK-2, FZ Jülich. Moreover, T. Vo's role as a teaching assistant in the