

Consistent Modeling and Numerical Simulation of Space-Charge Layer Formation in All-Solid-State Thin-Film Cells

Katharina Becker-Steinberger^{1,2*}, Stefanie Braun^{1,2}, Simon Schardt^{1,2}, Arnulf Latz^{1,2}

¹Helmholtz Institute Ulm
of Electrochemical Energy Storage
Helmholtzstr. 11, 89081 Ulm, Germany
{Katharina.Becker-Steinberger,Stefanie.Braun, Simon.Schardt, Arnulf.Latz}@dlr.de

²Institute of Engineering
German Aerospace Center
Pfaffenwaldring 9, 70569 Stuttgart, Germany
{Katharina.Becker-Steinberger,Stefanie.Braun, Simon.Schardt, Arnulf.Latz}@dlr.de

ABSTRACT

All-solid-state (ASS) batteries offer a promising opportunity to overcome the weaknesses of currently available battery technology. The use of non-flammable solid electrolytes (SEs) instead of liquid electrolytes enhances safety. In this contribution, we present a thermodynamic consistent model for ASS thin-film cells as well as first numerical simulation results of the transient processes within these cells. The multi-physical processes inside such cells are governed by the material properties of the utilized solids and the characteristics of the interfaces between the electrodes and the SEs. Particularly, the processes at the interfaces are considered to be responsible for the present limitations of ASS cells. When electrodes and electrolyte are brought together, charge is redistributed until local equilibrium is reached. The resulting space-charge layers increase the interface resistance and therefore worsen performance characteristics like the power density. A better understanding of these processes is essential, to improve the performance of ASS cells. Existing 1D models for ASS thin-film cells, apply Nernst-Planck (NP) equations or generalized NP equations, [1,2], to model the lithium-ion transport in the SEs. Our model is based on the recently derived "beyond NP" model for space-charge layer formation in SEs, [3]. It consists of several nonlinear partial differential equations (PDEs) in multiple domains as well as nonlinear inner boundary conditions (BCs). Generally, thermodynamic consistent models of multi-component systems of charged and uncharged constituents are based on the Maxwell equations, the law of mass, momentum, and energy conservation and are aligned with the 2nd law of thermodynamics. However, to limit the attention to processes on the relevant time scale, the laws are approximated quasi-statically. The system of PDEs valid in the bulk SE is a nonlinear, time-dependent drift-diffusion equation coupled with a Poisson equation and a quasi-static momentum equation. In contrast to NP models the lithium-ion transport within the SE is governed by one consistent flux containing contributions from all mobile species within the SE. Furthermore, a pressure correction is included. The laws of coupled thermo- and electrostatics give rise to jump conditions on the interfaces. Linking these jump conditions with a Butler-Volmer equation, [4], adjusted to the solids leads to nonlinear inner BCs. The system of PDEs is discretized by a purely implicit finite volume scheme. The resulting algebraic equation system is solved by a Newton-type method. Numerical examples indicate the improved physical correctness of the model. Computational results for different SE materials are presented and compared to experiments as well as known semi-analytical results [3]. Both material modifications and thickness reduction decrease the arising electric fields at the interfaces and therefore reduce the resistance. These insights give a useful perspective to improve ASS cells.

References

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