

Electro-chemo-mechanical behavior of lithium-ion battery electrodes

Ying Zhao*, Bai-Xiang Xu, Peter Stein

Department of materials science, Technical University of Darmstadt
Jovanka-Bontschits-Str. 2, 64287 Darmstadt
e-mail: {ying.zhao,xu,p.stein}@mfm.tu-darmstadt.de

ABSTRACT

Li-ion batteries are widely used in portable electronics and electric vehicles nowadays, since they have high energy densities and light-weighted designs. In pursuit of larger capacity, potential alloy materials such as Si, Sn, and Sb have been introduced as alternative anode materials to graphite. However, those electrodes experience irreversible mechanical degradation already after few charge/discharge cycles due to high stresses. Those stresses arise from changes in lattice dimensions and crystal structures, which are associated with overall volume changes and phase transformation.

In this presentation, a Cahn–Hilliard-type diffusion model coupled with large strains is formulated [1, 2]. Thereby the electrochemical reaction is modeled through a modified Butler–Volmer equation to account for the influence of the phase change and mechanical stresses on the reaction fronts. Moreover, concentration-dependency of the elastic moduli and the anisotropy of the diffusivity are also covered. The 3D numerical simulations are carried out by isogeometric finite element methods in order to treat the high order differential equations in a straightforward sense. Illustrative simulation examples are demonstrated to reveal the electrochemical reactions on particle surfaces and phase interfaces. Results show that the ratio between the timescale of reaction and the diffusion can have a significant influence on phase segregation behavior, so does the anisotropy of diffusivity. In turn, the distribution of the lithium concentration greatly influences the reaction on the surface, especially when the phase interfaces appear on exterior surfaces. The reaction rate increases considerably at phase interfaces, due to the large lithium concentration gradient. In the next step, we introduce a fracture order parameter in the phase-field formulation to incorporate the crack in the particle [2, 3]. Simulation results demonstrate that the segregation of a Li-rich and a Li-poor phase during delithiation can drive the cracks to propagate.

Acknowledgements. This work is supported by the “Excellence Initiative” of the German Federal and State Governments and the Graduate School of Computational Engineering at the Technische Universität Darmstadt. The author Xu would particularly like to thank the Adolf Messer Foundation for awarding her the Adolf Messer Price and for the financial support.

REFERENCES

- [1] Y. Zhao, P. Stein, and B.-X. Xu, Isogeometric analysis of mechanically coupled Cahn–Hilliard phase segregation in hyperelastic electrodes of Li-ion batteries. *Computer Methods in Applied Mechanics and Engineering*, vol. 297, pp. 325–347, 2015.
- [2] Y. Zhao, B.-X. Xu, P. Stein, and D. Gross, Phase-field study of electrochemical reactions at exterior and interior interfaces in Li-ion battery electrode particles, *Computer Methods in Applied Mechanics and Engineering*, in press.
- [3] B.-X. Xu, Y. Zhao, and P. Stein, Phase field modeling of electrochemically induced fracture in Li-ion battery with large deformation and phase segregation, *GAMM-Mitteilungen*, vol. 39, no. 1, pp. 92–109, 2016.

*Presenting author