Spatially resolved modeling and characterization of (de-)intercalation in Li-Ion battery materials

Joint DFG Project within SPP 1473 "WeNDeLIB"

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Abstract for partial project 11.4 (Prof. Steinbach)

Today, phase field simulations are well established in materials science for the investigation of complex mechanisms during phase transitions. For reviews, see ref. [1, 2]. The standard models comprise the thermodynamics of bulk and interfaces as well as the influence of diffusion and elastic mismatch. Special applications deal with magnetically or electrically driven phase transformations.[3,4] Few publications, however, are known in the field of electrochemistry.

Irreversible processes, including phase changes and localized stresses, during battery cycling lead to mechanical fatigue and thereby battery performance loss. The phase field method will be used to simulate charge transfer at the electrode-electrolyte interface, volume expansion during interdiffusion, phase transformation, and mechanical stress. The project is expected to give a complete and, for the first time, spatially and temporally resolved picture of the complex mechanisms of lithium transport and phase transitions in nanograined battery materials during (dis)charging. Using these results, a route to improved materials will be pinpointed.

The focus of this project lies on $Li_xFe[PO_4]$, representing a promising class of future low-cost electrode materials.[5] The possible device improvement gained by shrinking microstructural elements to the nanometer regime will be covered by including surface- and interface-related materials characteristics. To study the kinetics of the lithium ion flux, a novel finite interface dissipation model will be used:[6]

$$\phi_\alpha \dot{c}_\alpha = \vec{\nabla}\left(\phi_\alpha D_\alpha \vec{\nabla} c_\alpha\right) + P\phi_\alpha\phi_\beta\left(\tilde{\mu}_\beta - \tilde{\mu}_\alpha\right) + \phi_\alpha\dot{\phi}_\alpha\left(c_\beta - c_\alpha\right)$$

It introduces the so-called "permeability" (P in the above formula), a material constant which characterizes the ease with which atoms travel through the interface. Its relation to peculiarities of the electrode surface like the electrochemical double layer, adsorption and solvation processes, is currently under investigation.

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[1] I. Steinbach, "Phase-field models in materials science; a tutorial review", *Model. Simul. Mater. Sc.* **2009**, *17*, 073001.

[2] Y. Wang, J. Li, "Phase field modeling of defects and deformation", *Acta Mater.* **2010**, *58*, 1212-1235.

[3] A. Kazaryan, Y. Wang, Y. M. Jin, Y. U. Wang, A. G. Khachaturyan, L. Wang, D. E. Laughlin, "Development of magnetic domains in hard ferromagnetic thin films of polytwinned microstructure", *J. Appl. Phys.* **2002**, *92*, 7408-7414.

[4] L. Q. Chen, "Phase-field method of phase transitions/ domain structures in ferroelectric thin films: a review", *J. Am. Ceram. Soc.* **2008**, *91*, 1835-1844.

[5] A. K. Shukla, T. P. Kumar, "Materials for next-generation lithium batteries", *Curr. Sci. India* **2008**, 93, 314-331.

[6] I. Steinbach, L. Zhang, M. Plapp, "Phase-field model with finite interface dissipation", *Acta Mater.* **2012**, *60*, 2689–2701.