A Phase Field Model for Lithium Ion Battery Particles

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Introduction: We developed a 3-D COMSOL model of the ion intercalation of isotropic $LiFePO_4$ lithium ion battery particles based on Cahn-Hillard reaction kinetics. The model exhibited results similar to simple nonlinear diffusion or voltage plateauing and phase separation depending on the thermodynamic conditions as reflected by the enthalpy of mixing per site (Ω).

Computational Methods: Our model rephrases the fourth order Cahn-Hilliard equation in COMSOL's standard PDE format as a system of two fully coupled second-order partial differential equations in Li^+ concentration and bulk chemical potential respectively.

Results: Figure 1 shows single phase behavior for repulsive case Ω =-2.0 and voltage plateauing for 2 < Ω > 1. Figure 2 shows the sudden voltage changes associated with phase separation for Ω = 2.5 and Figure 3 depicts the ion concentration on the surface of the particle for Ω = 2.5.

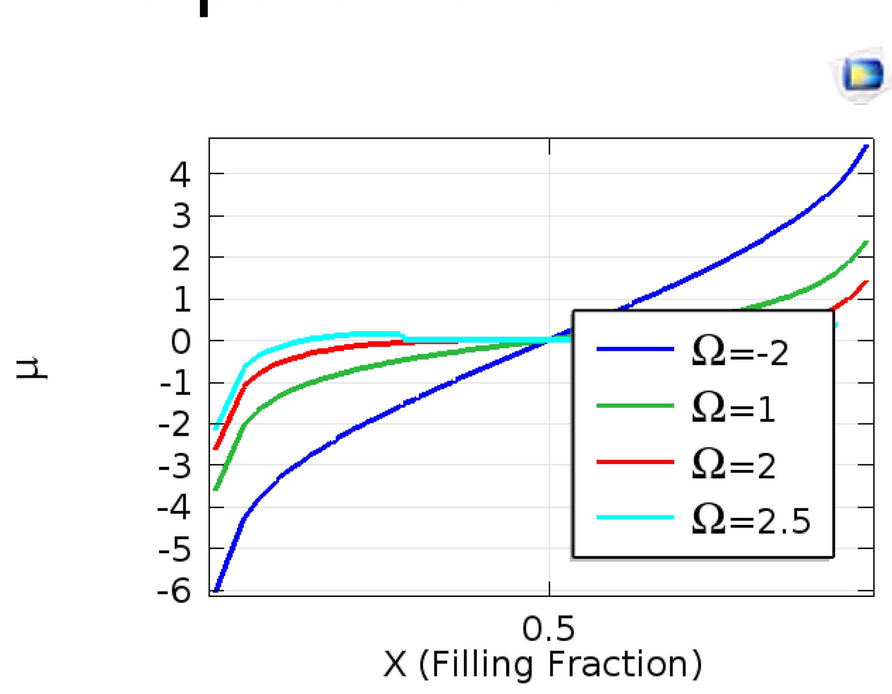

Figure 1: Voltage vs. filling fraction for single phase

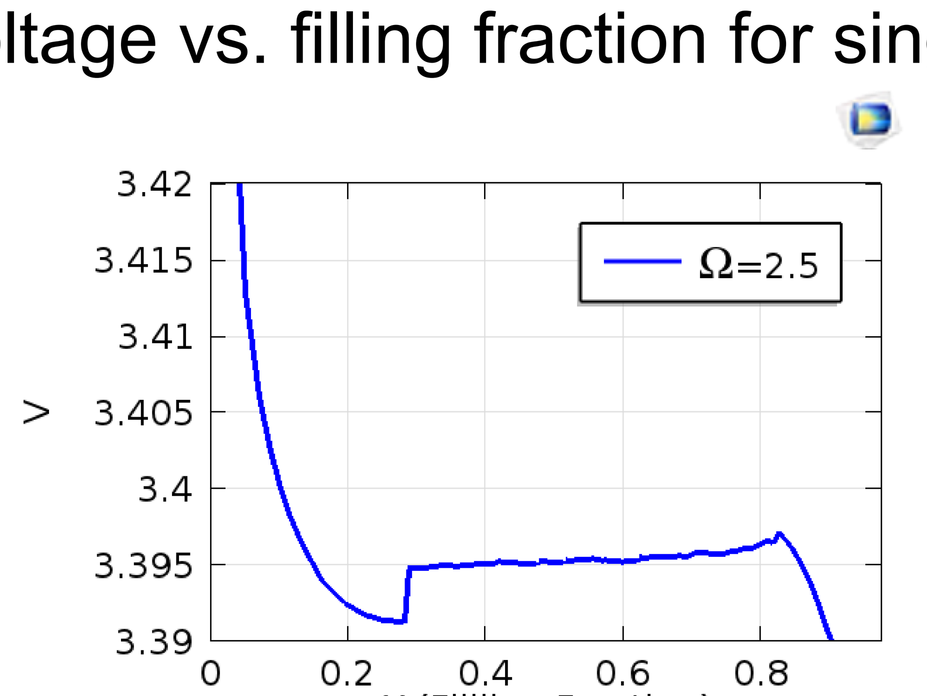

Figure 2: Two wave spinodal phase decomposition.

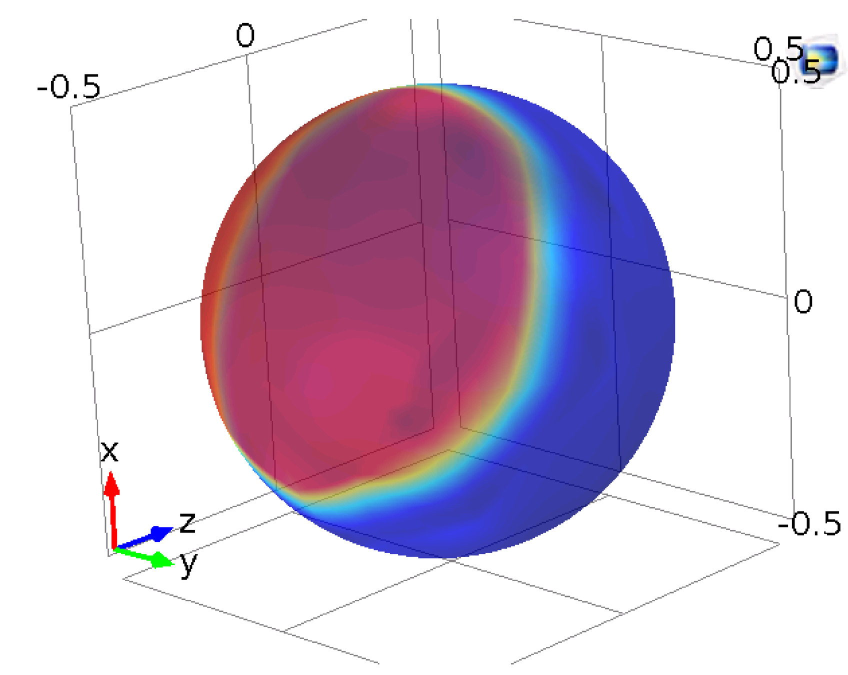

Figure 3: Ion concentration on the surface of the particle

Conclusions: This simple 3-D spherical Cahn-Hilliard Equation model is an improvement of a 1-D in radius model for visualization purposes. The major utility of our finite element model, though, is the ease of its adaptation to address more realistic particle geometry, anisotropy, and surface wetting. These improvements of our model will provide a robust simulation of the complex physics needed for calculations and interpretation of experimental results. One area that we plan to address is the impact of extreme temperature environments on the physics and more practically on the operation of $LiFePO_4$ batteries in extreme conditions.

References:

1. Zeng, Y. and Bazant M. Z., *Cahn-Hilliard reaction model for isotropic Li-ion battery nanoparticles*, MRS Proceedings, **1542**: (2013).
2. M. Z. Bazant, *Theory of chemical kinetics and charge transfer based on nonequilibrium thermodynamics*, Accounts of Chemical Research, **46 (5)**: pp 1144-1160 (2013).
3. White, L., White, R., Mathematical modelling of a Lithium Ion Battery, Proceedings COMSOL Conference 2009 Boston, (2009).
4. Painter, R., Berryhill, B., Sharpe, L., and Hargrove, S. K., *A Single Particle Thermal Model for Lithium Ion Batteries*, Proceedings COMSOL Conference 2014 Boston, (2014).
5. Singh, G., Ceder and Bazant, M., *Intercalation dynamics in rechargeable battery materials: General theory and phase trans-formation waves in $LiFePO_4$*, Electrochimica Acta 53, 7599-7613 (2008).