Towards optimal performance and in-depth understanding of spinel $Li_4Ti_5O_{12}$ electrodes through phase field modeling

Alexandros Vasileiadis [a], Niek J.J. de Klerk [a], Raymond B. Smith [b], Swapna Ganapathy [a], Peter Paul R. M. L. Harks [c], Martin Z. Bazant [b, d], Marnix Wagemaker [a]

[a] *Storage of Electrochemical Energy (SEE), Faculty of Applied Sciences, Delft University of Technology, The Netherlands.*

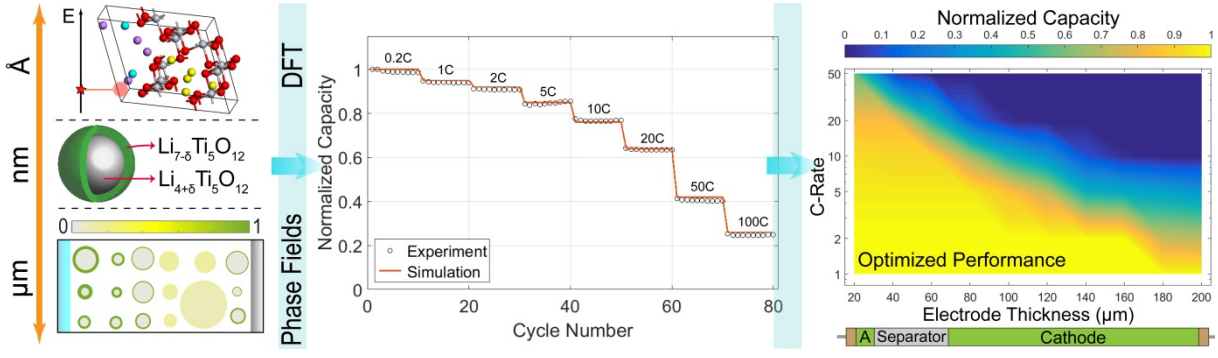
[b] *Department of Chemical Engineering, Massachusetts Institute of Technology, USA.*

[c] *Materials for Energy Conversion and Storage (MECS), Faculty of Applied Sciences, Delft University of Technology, The Netherlands.*

[d] *Department of Mathematics, Massachusetts Institute of Technology, USA.*

E-mail: A.Vasileiadis@tudelft.nl

Computational modeling is vital for the fundamental understanding of processes in Li-ion batteries. However, capturing nanoscopic to mesoscopic phase thermodynamics and kinetics in the solid electrode particles embedded in realistic electrode morphologies is challenging. In particular for electrode materials displaying a first order phase transition, such as $LiFePO_4$, graphite and spinel $Li_4Ti_5O_{12}$ (LTO), predicting the macroscopic electrochemical behaviour requires an accurate physical model. Herein, we present a thermodynamic phase field model for Li-ion insertion in spinel LTO. The model can capture the phase separating nature of the material via a regular solution description[1,2]. Phase stability between the Li-poor and Li-rich phases is based on ab-initio DFT calculations[3] and microscopy observations while the Li-ion diffusion parameters are based on nanoscopic NMR measurements[4]. The model successfully implements nanoscopic thermodynamic and kinetic properties to mesoscale, bridging the two length scales and eliminating the fitted, unknown parameters. Direct comparison with prepared electrodes shows good agreement over three orders of magnitude in the discharge current. Overpotentials associated with the various charge transport processes, as well as the active particle fraction relevant for local hotspots in batteries, are analysed in detail offering insight for optimal cycling. It is demonstrated which process limits the electrode performance under a variety of realistic conditions. These results provide concrete directions towards the design of optimally performing $Li_4Ti_5O_{12}$ electrodes. This is a step forward towards a consistent physical description of electrode materials for next generation modeling.



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