



# OXFORD BATTERY MODELLING SYMPOSIUM

## ONLINE PROGRAM

All times are BST

(British Summer Time daylight savings start on 28<sup>th</sup> March)

**Wednesday 31st March 2021**

<b>Time</b>	<b>Event</b>
14:15	Zoom session opens for people to try
14:30	<b>Gather and brief welcome</b>
14:45	<b>Daniel Steingart</b> – “Transport-Driven Chemical and Mechanical Behaviors in Closed Form Electrochemical Energy Storage”
15:35	<b>Yue Qi</b> – “Connecting Atomistic Simulations with Continuum Model for the Charge Transfer Reaction at the Electrode/Electrolyte Interface”
16:20	Break and Posters
17:00	<b>Birger Horstmann</b> – “Morphology and Function of SEI during Battery Operation”
17:45	Finish

We hope that you enjoy the afternoon and find the symposium inspiring and useful.

Organising committee of the Oxford Battery Modelling Symposium:  
David Howey & Charles Monroe (Department of Engineering Science), Jon Chapman &  
Colin Please (Mathematical Institute)

## Transport-Driven Chemical and Mechanical Behaviors in Closed Form Electrochemical Energy Storage

Daniel Steingart

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**Abstract:** The complex interplay between mechanics and chemistry at interfaces in batteries is receiving well deserved attention. Since these behaviors are not isolated to nanometer nor micron scales, and inserting probes to measure the behavior directly alters the behavior, capturing these couples is traditionally non trivial. In our work we exploit coupled length scales to put simple probes *outside* the cells because mechanical and chemical consequences of a given reaction reverberate through the entire cell.

The quest to maximize energy density only amplifies this coupling. Herein we consider the use of acoustic and optical measurements to inform the proficient modeler of some of the "long range" phenomena their models should capture to better describe dynamic behavior and degradation profiles of closed form systems. We will also venture to model some of these behaviors ourselves. Highlighted will be the acoustic response of lithium ion batteries, lithium metal batteries, and the optical response of passive flow ZnBr<sub>2</sub> batteries.

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**Biography:** Daniel Steingart is the Stanley Thompson Associate Professor of Chemical Metallurgy and Chemical Engineering and the co-director of the Columbia Electrochemical Energy Center. His group studies the systematic behaviors of material deposition, conversion, and dissolution in electrochemical reactors with a focus on energy storage devices. His current research looks to exploit traditional failure mechanisms and interactions in batteries, turning unwanted behaviors into beneficial mechanisms.

His efforts in this area over the last decade have been adopted by various industries and have led directly or indirectly to five electrochemical energy related startup companies, the latest being Feasible, an effort dedicated to exploiting the inherent acoustic responses of closed electrochemical systems. Steingart joined Columbia Engineering in 2019 from Princeton University where he was an associate professor in the department of mechanical and aerospace engineering and the Andlinger Center for Energy and the Environment. Earlier, he was an assistant professor in chemical engineering at the City College of the City University of New York. Even earlier he was an engineer at two energy related startups. He received his PhD from the University of California, Berkeley, in 2006.

## Connecting Atomistic Simulations with Continuum Model for the Charge Transfer Reaction at the Electrode/Electrolyte Interface

Yue Qi

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**Abstract:** The charge transfer reaction is the fundamental reaction for rechargeable batteries. The atomistic scale energy landscape of this reaction depicts the equilibrium and kinetics of the electrochemical process. Recently, we developed a density functional theory (DFT)-based electrode/electrolyte interface model to predict the macroscopic electrochemical equilibrium potential and the Butler-Volmer equation kinetics for electrodeposition reactions of  $M^{n+} + ne^- \leftrightarrow M^0$ .

This talk will review the basics of electrochemical processes and reveal the atomistic details of the solvation, desolvation, and charge transfer reactions at the Li and Mg electrode/electrolyte interfaces. For example, it was predicted that the Li-metal surface is negatively charged at the electrochemical equilibrium condition (the experimentally defined zero voltage for  $Li^+/Li^0$ ) due to the large solvation energy of  $Li^+$  in the EC-based electrolyte. The electric field created by the negatively charged surface can reorient the electrolyte into an ordered structure, lower the  $Li^+$  ion desolvation energy barrier, and help the  $Li^+$  ion transport through the SEI.

This atomistically predicted Butler-Volmer equation will enable a bottom-up multiscale modeling approach for Li-ion batteries. Here we demonstrate, with a DFT-informed phase-field model, the morphology difference between the dendritic Li plating and faceted Mg plating was captured.

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**Biography:** Dr. Yue Qi is the Joan Wernig Sorensen Professor of Engineering at Brown University. She received her Ph.D. degree in Materials Science with a minor in Computer Science from Caltech. She spent the next 12 years working at the General Motors R&D Center. At GM, she developed multi-scale models starting from the atomistic level to solve engineering problems related to lightweight alloys, fuel cells, and batteries. She transitioned from industry to academia in 2013 and served on the faculty in the Chemical Engineering and Materials Science Department at Michigan State University till 2020. Professor Qi and her “Materials Simulation for Clean Energy” Lab develop multi-scale simulation methods to couple chemical, electromechanical, point defects, transport, and mechanical phenomena and design materials and interfaces that are critically important for an energy-efficient and sustainable future. She has received several awards for her research, including the co-recipient of 1999 Feynman Prize in Nanotechnology for Theoretical Work for her Ph.D. work; three GM Campbell awards for fundamental research on various topics while working in GM; and the 2017 Minerals, Metals & Materials Society (TMS) Brimacombe Medalist Award for her contributions in multidisciplinary computational materials science. She is also passionate about increasing diversity in Engineering and served as the inaugurate Associate Dean for Inclusion and Diversity in the College of Engineering at MSU between 2018-2020.

## Morphology and Function of SEI during Battery Operation

Birger Horstmann

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**Abstract:** The solid-electrolyte interphase (SEI) on negative electrodes in lithium batteries is one of the main contributors to lithium-ion loss, capacity fade, and limited battery lifetime [1]. Our theory-based continuum models and their experimental validation give insights into the relevant transport and reaction mechanisms for electrons and lithium ions in the SEI.

Standard SEI models focus on capacity fade as a function of time. We developed a series of models to discuss additional dependencies, i.e., SEI morphology [2], potential dependence of SEI growth [3], dependence of SEI growth on cycling conditions [4], and lithium-ion concentration polarization inside SEI [5]. We gain novel insights into transport mechanisms by comparing with dedicated experiments, e.g., battery storage at various state-of-charge [6], differential capacity analysis during cycling [7], and impedance spectroscopy on model electrodes [8].

We highlight the importance of morphology for the function of SEI. It is often reported that SEI is more porous close to the electrolyte than close to the electrode. Our model can predict this dynamic transition in morphology by relating the intrinsic rates of SEI formation reaction and electron transport through the SEI.

On graphite electrodes, Jossen et al. measured the state-of-charge dependence of capacity fade [6]. We show how this reduces the set of potential transport mechanisms responsible for SEI growth [3]. We present the first indirect evidence that neutral radicals carry a negative charge and diffuse through the SEI. Bazant et al. have recently measured and simulated the differential capacity loss during cycling [4,7]. They observe that SEI growth during intercalation dominates. We extend our calendar-life model to describe the observed asymmetry in SEI growth during battery operation. We analyze with 3D micro-structure resolved simulations how the potential dependence of SEI growth results in SEI inhomogeneity throughout the negative electrode.

In this talk, we discuss a physics-based model for impedance spectroscopy of lithium batteries with SEI as porous surface film [5]. Validating our model with experiments of lithium metal electrodes [8], we find large transference numbers for lithium ions. This analysis reveals that lithium-ion transport through the SEI has solid electrolyte character.

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### References

- [1] B. Horstmann, F. Single, A. Latz, *Curr. Opin. Electrochem.* **2019**, *13*, 61–69, doi:10.1016/j.coelec.2018.10.013.
- [2] P. He, G. Zhang, X. Liao, M. Yan, X. Xu, Q. An, J. Liu, L. Mai, *Adv. Energy Mater.* **2018**, *8*, 1–6, doi:10.1002/aenm.201702463.
- [3] F. Single, A. Latz, B. Horstmann, *ChemSusChem* **2018**, *11*, 1950–1955, doi:10.1002/cssc.201800077.
- [4] S. Das, P. M. Attia, W. C. Chueh, M. Z. Bazant, *J. Electrochem. Soc.* **2019**, *166*, E107–E118, doi:10.1149/2.0241904jes.
- [5] F. Single, B. Horstmann, A. Latz, *J. Phys. Chem. C* **2019**, *123*, 27327–27343, doi:10.1021/acs.jpcc.9b07389.

- [6] P. Keil, S. F. Schuster, J. Wilhelm, J. Travi, A. Hauser, R. C. Karl, A. Jossen, *J. Electrochem. Soc.* **2016**, *163*, A1872–A1880, doi:10.1149/2.0411609jes.
- [7] P. M. Attia, S. Das, K. Lim, M. Z. Bazant, W. C. Chueh, *J. Electrochem. Soc.* **2019**, *166*, E97–E106, doi:10.1149/2.0231904jes.
- [8] F. Wohde, M. Balabajew, B. Roling, *J. Electrochem. Soc.* **2016**, *163*, A714–A721, doi:10.1149/2.0811605jes.
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**Biography:** Dr. Birger Horstmann is leading the team “Theory of Electrochemical Systems” at the German Aerospace Center in the Helmholtz Institute Ulm. He studied Physics and Mathematics at the University of Jena and the University of Cambridge. After performing his PhD on theoretical quantum physics at the Max-Planck-Institute for Quantum Optics near Munich, Dr. Horstmann moved to the field of continuum modelling of batteries. His current research focusses on mesoscale models for electrochemical interfaces and interphases. This includes the theory of electrochemical double layers, nucleation and growth of reaction products, and the formation of interphases, e.g. SEI. Furthermore, he evaluates these mesoscale theories in macroscopic models of lithium and beyond lithium batteries.