Simulation-based inference on metal plating dynamics

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Introduction

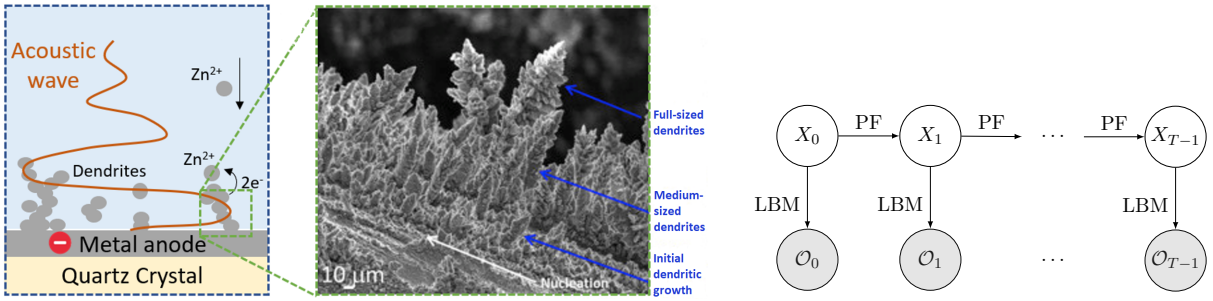
Realizing metal electrodes is a "Holy Grail" for rechargeable batteries. Metal electrodes offer not only superior energy, cost, and sustainability characteristics for existing battery systems, but they are also door-openers for alternative more sustainable chemistries [1]. Despite the recent surge in research, the inherent complexity of metal electrodes remains highly challenging. We aim to accelerate research on metal electrodes for energy storage by combining multiscale and multi-physics simulators with state-of-the-art techniques for statistical inference, thereby strengthening our ongoing collaboration on data-driven and high-throughput experimentation [8]. In more detail, we aim to formulate and numerically simulate a system of coupled PDEs that describe the multiscale, multi-physics, phenomena of metal plating, and to use simulation-based inference and differentiable programming to infer the underlying surface morphology from indirect measurements. The resulting insights and predictions will have important implications for the design and optimization of metal plating processes and will also provide a framework for understanding and solving other multiscale, multi-physics problems.

Chemistry Motivation

All batteries contain three essential components: a positive (cathode) and a negative (anode) electrode separated by an electrolyte. Electric energy is released by coupled electrochemical reactions involving a working ion M^{n+} at these electrodes, which during discharge generate a current flowing from the anode to the cathode via an outer circuit. Irrespective of chemistry (be it based on M = Li, Na, Zn, Al, Fe, etc.), the ultimate anode for a battery in terms of energy density, cost, processability, and sustainability is a metal electrode, which operates simply by plating M^{n+} and stripping the corresponding metal M during battery charge and discharge, respectively [1].

Despite their immense promise, metal electrodes in rechargeable batteries need to overcome major challenges before successful practical realization. In particular, as shown in Figure 1a, plating of metal during battery charge often leads to the formation of needle-like metal nanostructures on the electrode surface, called dendrites, which are brittle and break off as dead inactive metal that induces capacity loss. Even worse, dendrites can eventually pierce the barrier separating the anode and cathode, hence putting the battery at risk of shorting or exploding.

Metal plating is an inherently complex and highly dynamic process that depends on a multitude of physicochemical parameters. As the process only occurs during operation, a deeper understanding thereof must consider the metal electrode *as it operates* in the battery. Very few approaches offer such unique inside access, but Electrochemical Quartz Crystal Microbalance with Dissipation Monitoring (EQCM-D) stands out as it provides unique real-time access to key figures of merit that are otherwise inaccessible [5]. Berg's team has for several years developed a unique EQCM-D battery system, but to fully comprehend, and ultimately exploit the experimental data, requires accurate physicochemical modeling. Here, phase-field (PF) modeling has proven a highly promising simulation technique, not the least owing to its flexibility when it comes to incorporating effects at various scales [3]. By first simulating and generating metal dendrites with PF and then performing hydrodynamic simulations, a complete workflow simulating the EQCM-D response may be formed. For the hydrodynamic simulations of such mesoscopic systems, Lattice Boltzmann methods (LBM) are particularly well-suited. For this reason, Sjölund's team recently validated and extended an existing LBM implementation, and is currently rewriting it to support parallelization and automatic differentiation.



(a) (b)

Figure 1: **(a)** EQCM-D uses an acoustic wave to probe the plating and stripping of metal at the anode. Dendrites forming on the surface can seriously compromise performance and safety. **(b)** Probabilistic graphical model of the system: the surface morphology is the latent state X_t, the PF model defines the state transitions, and the LBM model gives the EQCM-D measurements $\mathcal{O}_t$.

Mathematics Motivation

Forward problem. Multiscale, multi-physics problems are ubiquitous in advanced real-world applications and are critical to understanding and solving many complex systems. Metal plating is a prime example of such a problem, which involves multiple, coupled, partial differential equations (PDEs) describing the electrochemistry and the hydrodynamics. We will use phase-field modeling to simulate dendrite formation. However, this forward model alone is not enough to fully understand the mechanisms for metal plating in real-world batteries. Ultimately, we are interested in the inverse problem: inferring the evolution of the surface morphology from EQCM-D experiments on operating batteries. However, EQCM-D only provides indirect measurements in the form of a (virtually instantaneous) frequency response to forced mechanical oscillations. This frequency response can be modeled by Lattice Boltzmann methods, but to connect its *evolution* to that of the surface morphology requires combining the LBM and PF models, which is challenging because these models act on widely separated timescales.

Multiscale modeling is burdened by computational inefficiency, especially with increasing complexity and scales; it is not uncommon to encounter "hidden" or unknown physics of interfaces, inhomogeneities, and other singularities. We will address this by emulating the phase-field model using a physics-informed neural network [4], which is a specific type of neural PDE solver that can exploit the variational formulation of phase-field models. Since the geometry is defined on a grid, it is natural to use a recurrent, or autoregressive, convolutional neural network. Such models are computationally efficient, in part because of their multilevel structure, and have the added advantage of allowing automatic differentiating. The latter is especially relevant since it would allow gradients to propagate through the combined PF+LBM simulator, allowing end-to-end optimization and learning.

Inverse problem. The inverse problem can be expressed as the Hidden Markov Model shown in Figure 1b. We do, however, face two key challenges. The first is that the model is only defined *implicitly* by the PF+LBM simulators; since the likelihood is inaccessible, standard inference techniques are not applicable. The second is the high dimensionality of the problem, especially since we aim to generate samples of the surface morphology consistent with the EQCM-D measurements.

The dominant approach for likelihood-free inference, Approximate Bayesian Computation (ABC), unfortunately scales poorly to high-dimensional data. A workaround is to design low-dimensional summary statistics, but these are often insufficient for high-quality inference. We will instead use modern techniques for simulation-based inference [2] leveraging gradient information from the PF+LBM models to dramatically improve sample efficiency. To generate realistic samples of the surface morphology, we will build on recently developed techniques for reducing dimensionality by parameterizing fields—surface morphology in our case—by a coordinate-dependent neural network [7]. Finally, we will encode physical knowledge as statistical priors, such that the resulting *Bayesian* inverse model enjoys the full range of advantages of the Bayesian machinery, including uncertainty quantification and automatic regularization. The inference could then be performed via likelihood-free variational inference [6].

Main Objectives

The forward and inverse problems each give rise to a number of more specific research goals:

Forward problem

- Define an accurate PF model of dendrite formation and validate it by experimental techniques that directly image the surface morphology (i.e. not EQCM-D).
- Couple the PF and LBM models to establish a multiscale model of the complete workflow.
- Improve computational efficiency of the combined model by introducing appropriate approximations and leveraging high-performance computing.

Inverse problem

- Define interpretable, low-dimensional, summary statistics of the surface morphology, and implement a baseline for likelihood-free inference, e.g. ABC. Verify that the LBM model can be used to infer morphology from EQCM-D measurements on a *static* morphology.
- Explore data-driven techniques for dimensionality reduction to replace the handcrafted descriptors of the surface morphology with more expressive ones.
- Adapt and extend existing techniques for simulation-based inference to obtain a—preferably Bayesian—model that can generate realistic samples of the surface morphology.

Interdisciplinary nature of the project. Mathematics is at the center of the project; the forward problem amounts to modeling and numerically solving partial differential equations, while the corresponding inverse problem relies on likelihood-free inference. Meanwhile, the project's overall scope is defined by the application to metal electrodes in rechargeable batteries.

Advisors. Jens Sjölund is a WASP assistant professor in AI with the mission to accelerate the progress of science. Sjölund has extensive experience developing methods for machine learning and optimization from industry and academia. Erik J. Berg is a professor of chemistry pushing the frontier on automation for high-throughput research on rechargeable batteries. Berg has pioneered the development of EQCM-D and is the main supervisor for two PhD students (starting 2021 and 2022) that are co-supervised by Sjölund and Broqvist. Peter Broqvist is an associate professor of chemistry working on multi-scale models and methods for computational materials chemistry with the aim to bring simulations closer to the complex, large-scale dynamical problems of the real world.

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References

[1] E. J. Berg et al. "Rechargeable batteries: grasping for the limits of chemistry". In: *Journal of The Electrochemical Society* 162.14 (2015), A2468.

[2] K. Cranmer, J. Brehmer, and G. Louppe. "The frontier of simulation-based inference". In: *Proceedings of the National Academy of Sciences* 117.48 (2020), pp. 30055–30062.

[3] A. Y. S. Eng et al. "Theory-guided experimental design in battery materials research". In: *Science advances* 8.19 (2022), eabm2422.

[4] S. Goswami et al. "Transfer learning enhanced physics informed neural network for phase-field modeling of fracture". In: *Theoretical and Applied Fracture Mechanics* 106 (2020), p. 102447.

[5] N. Shpigel, M. D. Levi, and D. Aurbach. "EQCM-D technique for complex mechanical characterization of energy storage electrodes: Background and practical guide". In: *Energy Storage Materials* 21 (2019).

[6] D. Tran, R. Ranganath, and D. Blei. "Hierarchical implicit models and likelihood-free variational inference". In: *Advances in Neural Information Processing Systems* 30 (2017).

[7] Y. Xie et al. "Neural fields in visual computing and beyond". In: *Computer Graphics Forum*. Vol. 41. 2. Wiley Online Library. 2022, pp. 641–676.

[8] J. T. Yik et al. "Automated electrolyte formulation and coin cell assembly for high-throughput lithium-ion battery research". In: *ChemRxiv* (2022). DOI: 10.26434/chemrxiv-2022-bstn2.