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Coupled Thermal-Electrochemical Full Battery Modeling of LiSi/FeS_2 Batteries

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Abstract: *Modeling a full thermal battery stack provides physical insights about axial cell variations that occur during start up, load variations, and freeze-out regimes that are not immediately apparent from single-cell models. In addition, a full battery model enables simulation of the electrical behavior of multi-tap batteries in combination with more complex thermal boundary conditions. A thermal model of a full battery can predict axial temperature variations in the battery, but the electrical performance of a multi-tap battery with a temperature gradient cannot be readily predicted by a single-cell model alone.*

Our prior work focused on development of a coupled multi-physics model for a single cell of a thermal battery [1] and simplification of that model to reduce computational cost [2]. In this work, we extend this approach to a two-dimensional full stack LiSi/FeS_2 thermal battery and focus on aspects of the full battery that are not predicted by the single-cell model. Since the batteries freeze from the outside in, the full battery model allows prediction of different lifetimes from different tap locations and can better simulate the battery performance under loads near the end of its life. We also demonstrate that the model can capture the effects of Joule heating and electrochemical reaction heating, which produces load-dependent spatially non-uniform effects that change the predicted battery lifetime. Sensitivity of the model predictions to model inputs was quantified using Sobol' indices, allowing relative effects of the various model inputs on predicted quantities to be compared.

Keywords: thermal battery; primary lithium cell; molten salt electrolyte; iron disulfide; computational modeling

Introduction

Our prior work on multi-physics modeling of thermally activated batteries has focused on single-cell models to keep computational times tractable and provide model validation against single-cell experimental data [1,3,4]. These efforts have been successful in terms of producing a model capturing a rich set of physical processes, including heat transfer, electrochemical reactions, ion and species transport, porous flow, capillary effects, phase change, and mechanical deformation. The model reductions applied in [1] have brought the computational cost of the model down enough to extend the model to full battery stacks of multiple cells.

In this work, we present a coupled modeling approach for a full battery, including multiple taps, thermal insulation, the ignition train, and the battery can. To make the model tractable, we separate the electrochemical stack from the thermal solve, which allows the use of different meshes for different physics with independent resolution requirements. We show that this model provides a substantial savings in computational cost while maintaining accuracy of key quantities of interest (QoIs) for a demonstration full battery.

The coupled modeling approach is demonstrated using an open-source battery [5]. Additionally, a sensitivity study was conducted on a structurally similar proprietary battery design using the same computational approach described in this work. Both electrical and thermal parameters are sampled simultaneously. This sensitivity study allows the exploration of the parameter space, demonstration of model robustness for different parameter inputs, and identification of important model parameters.

Methods

In this section, we briefly describe the full physics model and then outline the different approaches and unique challenges associated with extending the single cell model to a full battery.

Model: Many of the details of the current model have been described in our prior publications so we will focus on adaptations needed to extend the model to a full battery [1]. As shown in Figure 1, the model is separated into two domains–an electrochemical domain and a thermal domain. The energy equation is solved in the thermal domain and solid electrical conduction, species and ion transport in the molten electrolyte, electrochemical reactions, and porous flow of electrolyte are solved for in the electrical domain.

There is a two-way coupling between the two domains, with the thermal domain supplying temperature for the electrical domain, and the electrical domain providing a Joule heating energy source for the thermal domain. At each time step, the thermal domain is solved using the lagged Joule heating source, then the

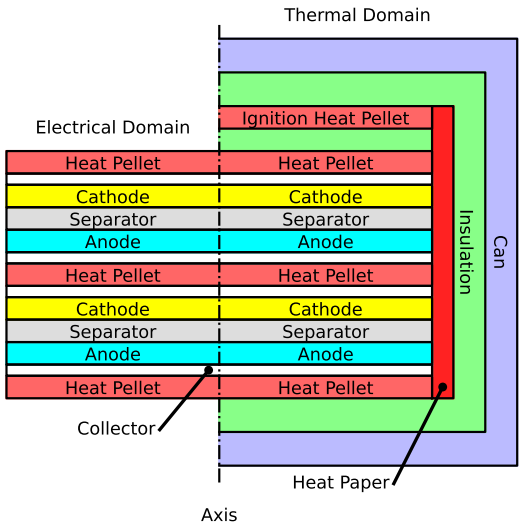

Figure 1. The two-dimensional axisymmetric simulation domains for a two-cell full-battery simulation (not to scale).

temperature field is interpolated into the electrical domain and the transport equations in the electrical domain are solved, then finally the updated Joule heating source is transferred back to the thermal domain.

The computational cost of the electrochemical domain is significantly higher than the thermal domain due to the larger number of equations, the expensive chemistry model evaluations, and the greater number of nonlinear iterations required to converge the more nonlinear set of equations. However, after the initial battery activation the electrochemical stack behavior is close to one-dimensional in the axial direction, while the thermal problem is largely radial through the insulation (except for the battery ends). For this reason, we can use a radially coarse mesh (10 elements) for the electrical domain while still using a radially finer mesh (100 elements) for the thermal domain to resolve the radial heat pellet burn progression and losses in the radial direction at later times. This approach provides considerable computational cost savings for a full battery, and allows better parallel load balancing for the electrical solve work than using a single domain.

Heat Pellet Ignition: The actual battery ignition is initiated at the center of the ignition heat pellet at the top of the stack (see Figure 1). This burns out to several discrete heat paper strips which burn down the length of the battery igniting the heat pellets in the stack. In this model, we do not model the heat paper burn, but instead initialize the level set field on all heat pellets to appropriately account for the heat paper burn speed.

The initial level set field (ϕ) in the ignition heat pellet

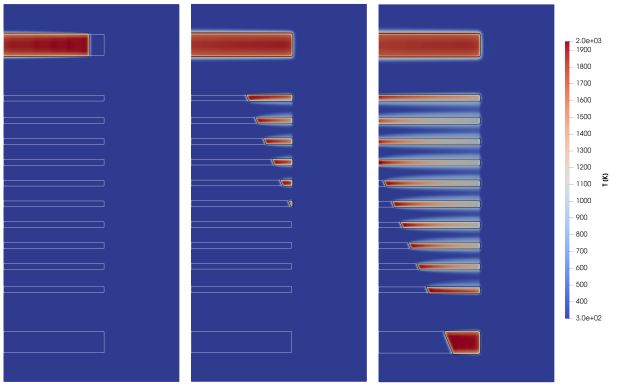

Figure 2. Simulated burn progression from ignition heat pellet down the stack to the remaining heat pellets. White lines show heat pellet locations and black lines show the position of the level set burn (and associated energy source).

is equal to the radius (r) since its burn initiates at the center and burns outward. On the remaining heat pellets, the level set field is initialized as

$$\phi = |z_{ign} - z| \frac{V_{pellet}}{V_{paper}} - r \tag{1}$$

where z_{ign} is the axial position of the ignition heat pellet, V_{pellet} is the burn speed of the heat pellet, and V_{paper} is the burn speed of the heat paper. This provides a realistic ignition timeline for the heat pellets without requiring resolution of the heat paper. Since the heat paper strips are not wrapped around the entire battery, accurately modeling them in a two-dimensional model would require additional adjustments to energy output and thermal properties.

The result of this initialization is a conical-shaped initial level set field that moves across the non-ignition heat pellets as shown in Figure 2.

Simulation Setup: The demonstration battery used for the simulations in this work is a 9-cell battery [5]. Electrical boundary conditions were applied at four collectors to provide a negative voltage tap (-2V), a 0V tap, and two positive voltage taps (nominally 4V and 14V, using 2 and 8 cells respectively). The time varying electrical load shown in Figure 3 was applied to all taps except the 0V tap, which used a fixed voltage boundary condition. After 1 second, the two positive taps have a steady high load, followed by a series of pulses over a lower base load. After the pulses end, the load is kept constant until the battery freezes out.

The exterior surface of the battery was set to a convective boundary condition to ambient temperature (300 K).

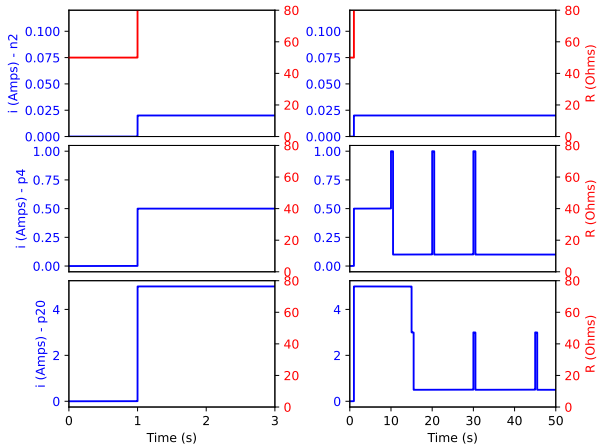

Figure 3. Battery loads (combination resistance and current loads) for all three battery taps. Loads are constant after 50 seconds. Current loads are in blue and resistive loads in red.

Sensitivity Studies: Sensitivity studies are an important part of model development which explore the parameter-to-QoI relationship. In the current study, we sample 55 parameters (both electrical and thermal) using Latin Hypercube Sampling (LHS), which is a method for efficiently and evenly sampling a high-dimensional space (for more details, see [6, 7]). Sensitivity results are summarized using Sobol' indices, which range between zero and one and measure the fraction of total QoI variance that can be attributed to the uncertainty in each parameter. For brevity, we refer the reader to [8, 9] for more details on Sobol' Indices. All parameters are sampled uniformly to avoid biasing the sensitivity results.

Results

The model described in the prior section was run from heat pellet initiation to freeze-out (the time where any cell is more than 90% re-solidified). To test the effect of the decoupling approach where the electrical domain uses a coarser mesh, the simulation was run in two configurations: one with a coarser electrical mesh (10 radial elements in the electrical mesh and 100 radial elements in the thermal mesh) and one with identical meshes (both using 100 radial elements). The effect on run-time of this change was substantial, with the different-mesh configuration taking 5.8x less time to complete the simulation on the same number of MPI ranks.

Voltage Predictions: The predicted voltage from all three taps is shown in Figure 4 for both mesh configurations.

The effect of multiple taps is visible in the 14 V tap trace at 10 and 20 seconds, when the current pulse on the 4 V tap causes a drop in the voltage supplied at the

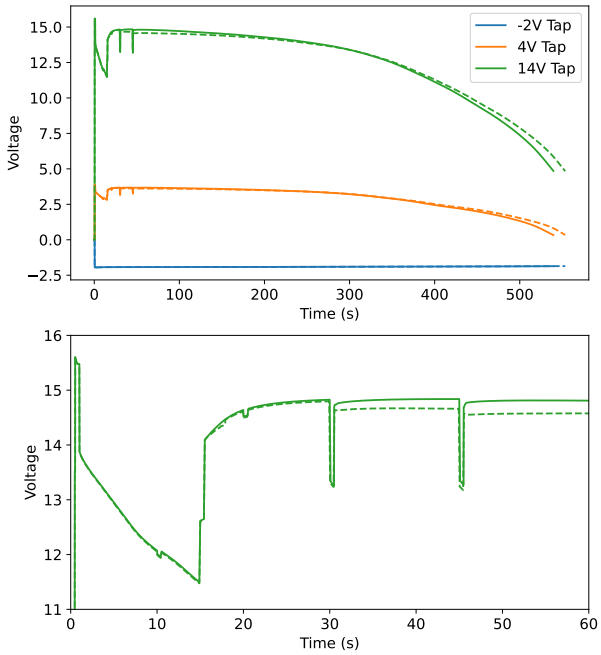

Figure 4. Predicted voltage vs. time with a coarser electrical mesh (solid lines) and identical meshes (dashed lines) showing performance under load and the relative effect of using a coarser electrical mesh.

14 V tap. The behavior with the different mesh configurations is nearly indistinguishable at early times, but there is a small difference in polarization loss in the 30 to 200 second range.

Some of the relevant quantities of interest (QOIs) from the full battery simulation are the rise time (defined here as time for the 14 V tap to reach 10 V), voltages at various times, and the freeze-out time (the time when any cell is more than 90% re-solidified). A comparison of how these QOIs change with the two mesh configurations is shown in Table 1.

Table 1. Change in quantities of interest from using dissimilar meshes (R10) vs the same mesh (R100)

QOI	R100	R10	Change
Rise Time (s)	0.457	0.455	0.44 %
V(10.2s)	11.967	11.987	0.17 %
V(25.0s)	14.756	14.781	0.17 %
V(60.0s)	14.576	14.809	1.60 %
V(200s)	14.190	14.281	0.64 %
Freeze (s)	553.45	539.49	2.52 %

Table 2. Main Sobol' Indices for simulation scalar responses (highlighted cells indicate $S > 0.10$)

Parameter	$T_{max,cath}$	$T_{max,ano}$	Rise Time	V_{max}
$V_{heatpellet}$	0.06	0.09	0.82	0.03
$k_{insulator}$	0.01	0.06	0.00	0.08
$k_{blanket}$	0.00	0.02	0.01	0.02
$C_{p,heatpellet}$	0.24	0.21	0.00	0.25
$C_{p,collector}$	0.17	0.13	0.00	0.27
$k_{cathode}$	0.35	0.02	0.00	0.03
$C_{p,insulator}$	0.07	0.03	0.00	0.03
$C_{p,anode}$	0.00	0.04	0.00	0.15
k_{anode}	0.01	0.30	0.00	0.00

Sensitivity Study: The Sobol' indicies for the various input parameters for select QOIs are shown in Table 2 (for a different battery). The most important input parameter for rise time prediction is the heat pellet burn speed ($V_{heatpellet}$). Other quantities, such as the maximum anode or cathode temperature, have a strong dependence on thermal properties (conductivity, k and specific heat, C_p). The predicted maximum voltage is sensitive to the specific heat of the stack components, likely through the temperature-dependence of the open-circuit voltage. These parameters, along with an uncertainty analysis, can help define which properties have the highest impact on predictions and which may require more detailed measurements.

Summary & Conclusions

In this work we presented a method for reducing the computational cost of a full-battery coupled thermal-electrical model using different mesh discretizations for different transport equations. We demonstrate that this approach provides greater than 5x reduction in computational cost while preserving accuracy of key quantities of interest.

We also developed a model sensitivity workflow to calculate Sobol' indices for a full-battery model to help quantify which model inputs have the largest impact on predicted quantities. This approach can be extended to include a large number of inputs and quantities of interest, and can be used to define where property measurements will provide the highest impact to the accuracy of the model predictions.

Acknowledgments

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