

ABSTRACT

The primary concern associated with lithium-ion batteries is safety during high-power utilization. Being able to estimate the internal states of the battery accurately in real-time is essential to prevent unsafe operating conditions. Furthermore, understanding the heat generation phenomenon during the cycling of cells is required as using these cells in fast-charging and high-power applications will become ubiquitous as the world shifts towards renewable energy generation. Towards this goal, the present study proposes a reduced-order electrochemical model that can be implemented for state estimation in real-time algorithms. Moreover, the heat of mixing in lithium-ion cells is modelled using a volume-averaged local thermal model, and its significance towards heat generation in lithium-ion cells is analyzed. Lastly, the proposed thermo-electrochemical model is simulated to predict the temperature of cylindrical lithium-ion cells. Its results are validated against experimental data. Various parameters required in the thermal and electrochemical models are experimentally determined.

Reduced-order physics-based models can achieve accurate state estimation due to the implementation of the inherent physics while remaining computationally inexpensive to be applied in real-time estimation-based algorithms. Towards this goal, a polynomial-based isothermal reduced order model is proposed in this work. This model can account for the spatial and temporal dynamics of the electrodes and the electrolyte. A novel approach is developed to account for the spatially varying overpotential and open-circuit potential. Moreover, we employ a semi-analytical model for lithium diffusion in the electrode and exhibit that it can provide great flexibility between accuracy and computational cost. We provide a criterion for truncating the semi-analytical solution to achieve high accuracy under fluctuating current applications based on the electrodes' characteristic diffusion time scale. A comparative analysis performed with an existing reduced-order polynomial model demonstrates that our model reduces error by a factor of five during cell voltage prediction for currents as high as $7C$. The performance of our

model is also presented under dynamic test conditions like US06 and FTP75 drive cycles. The computational time taken by our solver is close to the single-particle model despite incorporating electrolyte dynamics.

Moreover, we propose a local thermo-electrochemical pseudo-two-dimensional model accounting for the heat of mixing in the electrodes. Due to a lack of research towards the heat of mixing, the understanding of heat of mixing is fairly limited for lithium-ion cells. To the best of our knowledge, such a model has been implemented for the first time. The model is used to simulate, analyze and understand the temporal and spatial variation of heat generation in a lithium-ion cell. We show that the heat of mixing can contribute significantly towards the total heat generation in the cell under poor diffusion dynamics conditions and should be considered when developing a thermal model. In our simulations, the heat of mixing contributed as high as 9%-16% for 1C-5C discharge current and 38%-48% for 2C-5C charge current towards the total heat. In contrast to an earlier study, we show that it is not necessary for the contribution of the heat of mixing to increase with the current rate. Several sources like the ohmic heat, irreversible heat of reaction, and the heat of mixing increase with current, and the net interplay between these sources decide which component will become more significant at higher currents. Finally, a reduced-order formulation of this model is validated against the experimental cell voltage and temperature data for cylindrical cells. Experimental determination of several thermodynamical, electrochemical and geometrical model parameters is carried out. The dimensions of the cylindrical cell components are determined by disassembling the cell. Material characterization techniques, including scanning electron microscopy and energy-dispersive x-ray spectroscopy, are performed to determine the electrode composition and particle sizes. The galvanostatic intermittent titration technique is employed to obtain the diffusion coefficient of the electrode materials.