Analytical description of open circuit voltage influences on the current density distribution in lithium-ion cells

Andreas Noel [a], Franz B. Spingler [a], Devin Atukalp [a], Simon V. Erhard [a], Andreas Jossen [a]
[a] *Technical University of Munich (TUM), Institute for Electrical Energy Storage Technology (EES), Arcisstrasse 21, 80333 Munich, Germany*

E-mail: *andreas.noel@tum.de*

Fast and accurate characterization methods for lithium-ion cells are required in both stationary and automotive applications. Commonly used characterization methods, e.g. electrochemical impedance spectroscopy, require a steady state in the cell. Therefore, sufficient time for relaxation needs to be included in the measurement procedures, which compromises measurement speed [1]. Understanding the origin of and quantifying charge inhomogeneities and the corresponding relaxation processes is essential to design fast and accurate measurement methods.

During operation, current density distribution and potential drop along the current collectors are non-uniform, depending on the prevailing, local state of charge (SOC) [2-5]. The origin of an inhomogeneous current density distribution can be traced back to the ohmic potential drop along the current collectors. A linear open circuit voltage (OCV) results in a linear current density distribution along the electrode. With variations in the gradient of the OCV characteristic, however, the current density distribution is dynamic, nonlinear and non-monotonic over the SOC.

For further investigating these effects, a highly reduced model is presented that can be parameterized exclusively by full cell measurement data and is valid for a specific operating window. This model is based on only three components: A conductivity representing a segment of the current collector, a fundamental polarisation parameter representing the electrochemical system [6, 7] and a nonlinear capacitance that defines the change of voltage over charge analytically. A hyperbolic tangent expression was used to describe a well-defined change in voltage and differential voltage with charge, resembling the voltage characteristics of the LiC_6/LiC_{12} phase change in the graphite electrode. The model was validated against experimental data from a multi-tab-cell [2].

The results show a strong correlation between the second derivative of voltage over charge and the dynamics of current density distribution along the current collector. Additionally, the study proofs that a constant voltage gradient over charge generates a homogeneous current density distribution and a constant SOC difference along the electrode. These results permit designing ideal conditioning methods that reduce relaxation times and, hence, allow for faster and more accurate characterization measurements.

References:

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