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Title: Pseudo-Two-Dimensional Modeling of Lithium-Ion Conversion Cathode Materials

Authors: Jeffrey S. Horner, Grace Whang, Igor V. Kolesnichenko, Bruce S. Dunn, Timothy N. Lambert, and Scott A. Roberts

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Abstract:

Conversion cathode materials are an intriguing next-generation chemistry for lithium-ion batteries due to their high theoretical capacity. Nevertheless, there are various practical issues inhibiting current use in consumer products including limited rechargeability due to capacity fade and poor performance at fast cycling rates. Partially responsible for these issues is the current lack of understanding of leading loss mechanisms for conversion cathode materials as the changing composition of the electrode makes it difficult to isolate and eradicate cell limitations. Electrochemical models are typically used in combination with experiments to improve understanding of cell performance and limitations. However, in comparison to intercalation cathode materials, fewer modeling studies have been conducted for conversion cathode materials. As such, a robust and efficient model for lithium-ion conversion cathode electrochemistry is desirable to optimize these materials for practical use.

Here, we present a pseudo-two-dimensional model for conversion cathode materials implemented using the open-source Python Battery Mathematical Modelling (PyBaMM) software. We apply the model to FeS_2 – an intriguing material due to its abundance and high theoretical capacity. Upon discharge, FeS_2 first undergoes an intercalation reaction followed by a conversion reaction with lithium to produce Li_2S and Fe. The model is based on a polydisperse, core-shell approach for active material particles where the intercalated FeS_2 core converts to a shell of Li_2S, and two distinct sources for the current associated with the two separate reactions are enabled at the reaction surface. Upon initiation of the conversion reaction, an additional parameter representing the thickness of the converted shell material is tracked and used to incorporate ionic and electronic transport losses through this material. By comparing the model with experimental discharge data, we identify the leading loss mechanisms as poor solid-state diffusion during the intercalation reaction and poor electronic conductivity through the converted shell during the conversion reaction. Our analysis helps to guide design and optimization of FeS_2 cathodes, and the adaptability the model encourages its use for other conversion cathode materials.

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