Modelling Poster Abstract

Improving the performance of Li-S cells, a model-informed approach

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Lithium-sulfur (Li-S) could provide the next step-change in battery technology with a promising practical energy density of 500-600 Wh/kg, increased safety and possible low cost. However, a lack of understanding of the complex electrochemical, transport, and phase-change phenomena in Li-S cells is arguably holding back improvements in their performance1 and thus its successful commercialisation. Acquiring this knowledge requires experimental characterisation in tandem with mechanistic modelling.

To address the latter, we developed a zero dimensional model that captures the essential features of Li-S performance during charge and discharge. The model accounts for two electrochemical reactions via the Nernst formulation, power limitations through Butler–Volmer kinetics, and precipitation/dissolution of low order polysulfides, including nucleation.[2] The model is an improvement on the existing zero dimensional models, while requiring considerably fewer input parameters and computational resources than one dimensional models. Model results help identify the dominant effects in a real cell.[3] The flat shape of the low voltage plateau typical of the lithium–sulfur cell discharge is caused by precipitation. During charge, it is predicted that the dissolution can act as a bottleneck, resulting in reduced charge capacity and an earlier onset of the high plateau reaction, and indicated by the merging of the two voltage plateaus.

Here we use this model as a base to understand the interplay between shuttle, shuttle-related degradation and precipitation, and the impact on the total capacity available from a cell throughout its lifetime. To this end, model predictions of cell performance under cycling are compared to experimental data gathered for various operational conditions. The model helps interpret the complex behaviour observed during cycling and quantify two types of degradation: reversible and irreversible. The model interprets the observed SOC drift during cycling as mainly caused by accumulated precipitation (reversible loss), while showing that some features can only be obtained if active material is gradually lost irreversibly. The prediction of a reversible loss associated with precipitation is verified experimentally by the successful use of 'recovery' cycles.

This model can thus be used to make informed choices regarding improving performance while sacrificing either power or energy, such as through the introduction of recovery cycles.

References

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