4D Microscale Modelling of Lithium-Ion Battery Performance: SEI Growth and Data-Driven Surrogate Modelling

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Lithium-ion batteries (LIBs) are the leading technology in portable energy storage due to their high energy and power density. Despite their widespread applications, long-term performance remains limited by ageing phenomena, notably the growth of the Solid Electrolyte Interphase (SEI) on the anode surface. Formed through complex side reactions during early cycling, the SEI stabilizes the electrode–electrolyte interface. However, its structural inhomogeneity and instability often result in continued lithium consumption and progressive capacity loss. The experimental characterization of SEI remains challenging due to its nanoscale dimensions, chemical sensitivity, and compositional variability, highlighting the importance of computational modelling in advancing the understanding and control of this process.

This work presents a time-dependent modelling approach that bridges microscale electrochemical behaviour and macroscale cell performance. A four-dimensional (3D spatial + time) finite element model developed in COMSOL Multiphysics is employed to simulate SEI evolution during the initial cycles of a half-cell. By resolving the geometry of the porous electrode, the model captures local variations in lithium concentration and electrical potential, revealing spatial patterns in SEI growth. These insights highlight the importance of microstructural heterogeneity in determining degradation pathways, an aspect often overlooked in continuum-scale models that assume homogeneous electrode properties.

To address the computational cost associated with high-fidelity simulations, the second part of this study explores a data-driven surrogate modelling approach. Convolutional Neural Networks (CNNs) are trained to predict electrochemical behaviour based on 3D electrode geometries, offering a computationally efficient alternative for analyzing structure–performance relationships. The training set is obtained from physics-based simulations that exclude SEI formation but systematically vary the cathodic active material (different Nickel-Manganese-Cobalt oxides) and particle packing arrangement. The model is then validated using cathode tomographies to assess its predictive capability on realistic microstructures.

By combining physics-based and machine learning methods, this work aims to improve the performance and longevity of current energy storage technologies.

Keywords: *Lithium-ion battery, Solid-Electrolyte Interface (SEI), 4D Model.*