Novel quantification techniques for electrolyte motion induced salt inhomogeneity in aged battery electrodes

Mahla Bakhshi[1], Nour Adrah[2], Prof Johannes Wandt[1,3], Prof Johannes Landesfeind[1], Yuqing Zhao[4], Parham Taghizadegan[5], Michel Johnsen[4], Prof Michael Metzger[4], Prof Jeff Dahn[4,5]
[1]Department of Engineering Sciences, University of Agder, Grimstad, , Norway, [2]Department of Engineering and Design, Chair of Electrical Energy Storage, Technical University of Munich , , Germany, [3]BMW group, Munich, , Germany, [4]Department of Physics and Atmospheric Science, Dalhousie University, Halifax, , Canada, [5]Department of Mechanical Engineering, Dalhousie University, Halifax, , Canada

Electrolyte motion induced salt inhomogeneity (EMSI) has recently emerged as an important degradation mechanism in commercial lithium-ion batteries under fast-charging conditions.[1] During cycling, electrolyte redistribution can create salt concentration gradients across the cell, leading to local variations in ionic conductivity, and degradation behavior. However, directly measuring these inhomogeneities remains challenging due to the limited spatial resolution and complexity of existing techniques.

In this work, two complementary methods are presented to investigate and quantify EMSI in commercial lithium-ion batteries. First, an in-house developed impedance-based technique was used to locally quantify electrolyte salt concentration in extracted electrodes using dilution of residual salt and a custom multi-wire impedance probe. Second, an ex-situ micro X-ray fluorescence (μ-XRF) method was developed to directly map salt distributions across full electrodes by using $LiAsF_6$ as a tracer salt and calibrating the arsenic signal to local salt concentration.
Both methods revealed pronounced centimeter-scale salt gradients and tab-correlated inhomogeneities during cycling, demonstrating that EMSI in large-format cells is significantly more spatially heterogeneous and more strongly coupled to local pressure inhomogeneities than previously recognized, with important implications for commercial cell design and fast-charging strategies.

References

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