How multiscale modelling can help understand instabilities in the battery's microscale processes

Lithium based batteries are considered for many potential next-generation batteries, employing lithium metal anodes [1]. The performance and durability of lithium batteries are largely influenced by the operating conditions and often find their underlying cause in the nano- and microscale physical processes at the interfaces of the negative electrode. The solid electrolyte interphase (SEI) forms both on graphite and lithium metal, as the potential of the anode is lower than the reduction potential of the electrolyte. The SEI is known to be both a vital part of the battery and a key factor of contributing to its ageing. Although the capacity-fade due to continued SEI growth [2] and, in the case of lithium metal anodes, trapped dead lithium inside the SEI [3] are experimentally well known, fundamental processes are not fully understood and are still under debate.

We develop physically motivated mesoscale models to explain observed instabilities on the negative electrode and investigate its dependence on operating conditions. For the continued SEI growth on graphite, we present a surface growth model [4] combined with a diffusion-based SEI growth mechanism [5]. We observe a universal instability that can explain the emergence of a dual-layer SEI with an inner compact layer and an outer porous layer. For lithium metal anodes, we develop a generalized phase-field model [6] of the dissolution of a single lithium whisker covered by SEI. We find that an instability caused by the attraction between lithium metal and SEI leads to the formation of dead lithium.

1. Werres, M., Horstmann, B. *et al.* Strategies towards enabling lithium metal in batteries: Interphases and electrodes. *Energy Environ. Sci.* **14**, 5289–5314 (2021) doi:10.1039/d1ee00767j.
2. Keil, P. *et al.* Calendar Aging of Lithium-Ion Batteries. *J. Electrochem. Soc.* **163**, A1872–A1880 (2016) doi:10.1149/2.0411609jes.
3. Fang, C. *et al.* Quantifying inactive lithium in lithium metal batteries. *Nature* **572**, 511–515 (2019) doi:10.1038/s41586-019-1481-z.
4. Horstmann, B. *et al.* Rate-Dependent Morphology of Li 2 O 2 Growth in Li–O2 Batteries. *J. Phys. Chem. Lett.* **4**, 4217–4222 (2013) doi:10.1039/c3ee40998h.
5. Kolzenberg, L., Latz, A. & Horstmann, B. Solid–Electrolyte Interphase During Battery Cycling: Theory of Growth Regimes. *ChemSusChem* **13**, 3901–3910 (2020) doi:10.1002/cssc.202000867.
6. Bazant, M. Z. Theory of chemical kinetics and charge transfer based on nonequilibrium thermodynamics. *Acc. Chem. Res.* **46**, 1144–1160 (2013) doi:10.1021/ar300145c.