Strategies to Spatially Guide Li Deposition in Porous Electrodes for High-Performance Lithium Metal Batteries

Janina Drews,[1,2] Syed Abdul Ahad,[3] Hugh Geaney,[3] Timo Danner[1,2] and Arnulf Latz[1,2,3]
[1] *German Aerospace Center (DLR), Institute of Engineering Thermodynamics, Ulm, Germany*
[2] *Helmholtz Institute Ulm (HIU), Ulm, Germany*
[3] *University of Limerick, Department of Chemical Sciences and Bernal Institute, Limerick, Ireland*
[4] *University of Ulm, Institute of Electrochemistry, Ulm, Germany*
Wilhelm-Runge-Straße 10, 89081 Ulm, Germany
janina.drews@dlr.de

To exploit the high theoretical energy density of Li metal anodes, critical issues like drastic volume changes and dendritic Li growth need to be overcome. A rational design of the anode is key to enhance the safety and extend the cycle life of Li metal batteries. Thereby, 3D host architectures are a promising strategy to alleviate volume changes. However, it is challenging to ensure a bottom-up infilling and prevent Li depostion and consequent dendrite formation on top of the host structure.
In our contribution we carefully study the impact of structural and physical properties of the host matrix on Li deposition. Therefore, we present an extended continuum model,[1,2] which explicitly considers substrate characteristic barriers for Li nucleation. The model is implemented in a 1D as well as in 3D framework, whereby former additionally includes changes of electrolyte and Li volume fraction and thereby caused changes in the specific surface area available for Li plating. The parameterization and validation of the model is based on literature data and dedicated experiments.
Extensive parameter studies - including numerous 3D microstructures[3] - reveal key design strategies to achieve the desired bottom-up filling of the porous host. Overall, such an infilling is only observed for hosts with a very low electric conductivity or when preferred nucleation sites are introduced by a lithiophilic interlayer, i.e. Ge (Figure 1), while the microstructure on its own can reduce but not prevent the dominating on-top plating. Thereby, the simulations provide a roadmap to effectively design porous host architectures which enable spatially controlled Li deposition.

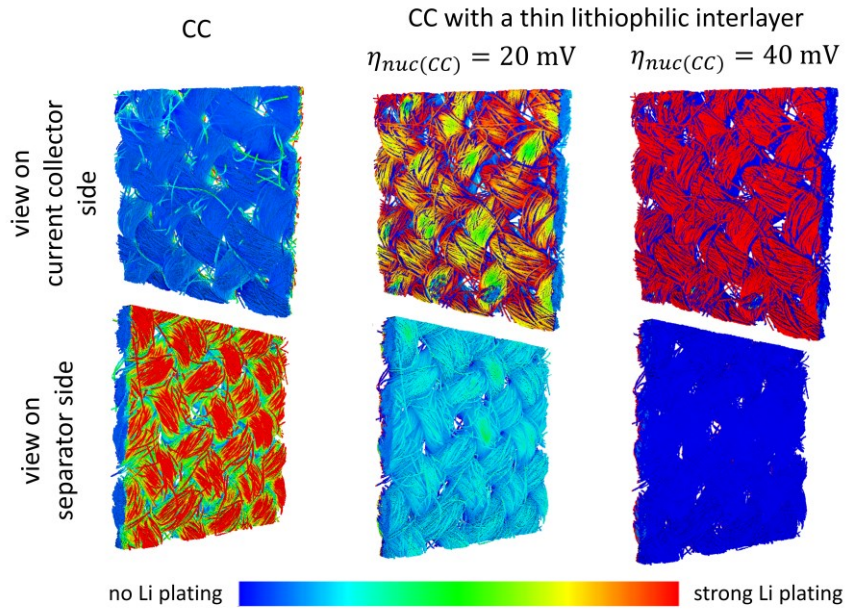

Figure 1 Amount of plated Li on carbon cloth (CC) with and without a lithiophilic interlayer after 100s at 0.1 mAcm^{-2} and overpotentials for Li nucleation on CC of $\eta_{nuc(CC)}$ = 20 / 40 mV.

[1] A. Latz and J. Zausch, *Journal of Power Sources*, **2011**, *196*, 3296-3302.
[2] S. Hein, T. Danner A. Latz, *ACS Applied Energy Materials*, **2020**, *3*, 8519-8531.
[3] B. Prifling, M. Röding, P. Townsend, et al., *Frontiers in Materials*, **2021**, *8*, 786502.