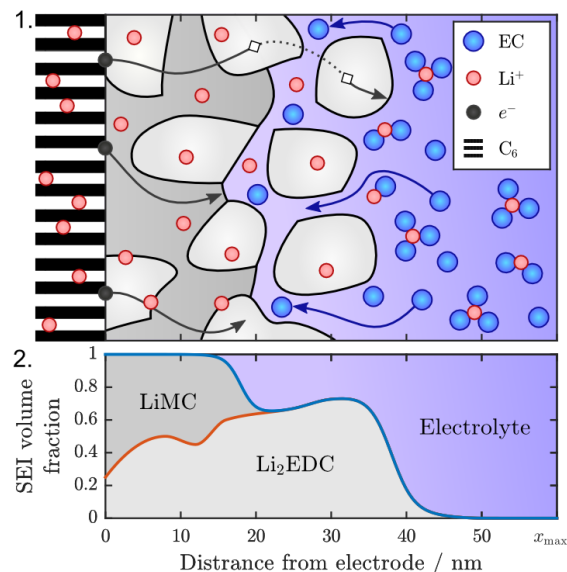


Abstract for oral presentation**Theory-based Investigation of SEI Formation****Fabian Single^{*1,2}, Birger Horstmann^{1,2}, Arnulf Latz^{1,2,3}**¹ German Aerospace Center (DLR), Institute of Engineering Thermodynamics, Stuttgart, Germany² Helmholtz Institute Ulm (HIU), Ulm, Germany³ University of Ulm, Institute of Electrochemistry, Ulm, Germany

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The solid electrolyte interphase (SEI) is a thin film which separates the electrolyte from an electrode, e.g. the negative electrode in lithium-ion batteries. It is composed of electrolyte reduction products which are rapidly generated as long as the electrolyte is in contact with a negatively polarized and pristine electrode. This occurs during the first charge of the battery where SEI is quickly formed [1]. The SEI is passivating for electrons as well as electrolyte molecules and therefore stops the rapid electrolyte reduction. However, slow, long-term SEI formation reveals that electrolyte reduction is not completely suppressed by this passivating layer. Therefore, at least one SEI precursor can migrate across the SEI at a small rate. Now, the throughput of this transport processes limits the rate of long-term SEI formation. Our model [2] uses this assumption to describe the evolution of a porous SEI as sketched in Figure 1. Our approach is similar to previous SEI model studies [3,4]. However, we trace the evolution of SEI volume fraction along the axis perpendicular to the electrode surface as shown in Fig. 2. This results in novel predictions, e.g., morphological properties such as the dual-layer structure.

The transport of SEI precursors, i.e., electrons, electrolyte molecules or neutral lithium interstitials [5], through either the SEI or its pores can be adjusted in our model. Theoretically, each of these processes can facilitate SEI growth on its own. Therefore, we investigate SEI formation for each these transport mechanisms individually. In this way, we predict how the corresponding films can be distinguished. They do not only differ in the potential dependence of their formation rate but also in the dual layer structure. Additionally some mechanisms tend form SEI with homogeneous film thickness whereas others form SEI with strongly fluctuating thickness. These results allow experimentalists to do draw conclusions with respect to the actual rate-limiting transport process, if they observe one of these features in a suitable experiment.



- [1] Verma, P., Maire, P., & Novák, P. (2010). *Electrochimica Acta*, 55(22), 6332–6341. doi:10.1016/j.electacta.2010.05.072
- [2] Single, F., Horstmann, B., & Latz, A. (2016). *Phys. Chem. Chem. Phys.*, 18, 17810–17814. doi:10.1039/C6CP02816K
- [3] Pinson, M. B., & Bazant, M. Z. (2012). *Journal of the Electrochemical Society*, 160(2), A243–A250. doi:10.1149/2.044302jes
- [4] Ploehn, H. J., Ramadass, P., & White, R. E. (2004). *Journal of The Electrochemical Society*, 151(3), A456. doi:10.1149/1.1644601
- [5] Shi, S., Lu, P., Liu, Z., Qi, Y., Hector, L. G., Li, H., & Harris, S. J. (2012). *Journal of the American Chemical Society*, 134(37), 15476–15487. doi:10.1021/ja305366r