# Solvation Behaviour in Electrochemical Double Layers: Modelling Solvation Energy

C. Schwetlick[1,2,4]*, M. Schammer[2,3], B. Horstmann[1,2,3], A. Latz[1,2,3,4]

[1]Universität Ulm, Albert-Einstein-Allee 47, 89081 Ulm, Germany
[2]Institute for Technical Thermodynamics, DLR, Pfaffenwaldring 38-40,
70569 Stuttgart, Germany
[3]Helmholtz Institute Ulm, Helmholtzstraße 11, 89081 Ulm, Germany
[4]POLiS Cluster, Lise-Meitner-Straße 16, 89081 Ulm, Germany
** email address: constantin.schwetlick@dlr.de*

Electrolyte modelling promises to speed up processes surrounding design and selection of electrolytes for batteries [1]. Models which can accurately describe dilute electrolytes are known for some time. Highly concentrated electrolytes, however, are more difficult to model. Solvation mechanics can play a very important role in the battery performance, for example when comparing the intercalation capacity of graphite electrodes regarding Li and Na-Ions [2]. Therefore, the desolvation inside the double layer plays a central role in the performance of graphite electrodes [3]. For this reason, including solvation behaviour in the model makes it applicable to a much wider range of batteries.

Dreyer et al. [4] presented a model for electrolytes which includes solvation effects. The model assumes a dilute solution and fixes the number of solvent molecules bound to each ion. We have recently presented a theory for highly concentrated electrolytes based on a free energy which includes electrostatic energy, the contributions of the molar volumes of the species and the entropic energy [5,6]. In this contribution, we propose to add an interaction energy for ion solvation in concentrated electrolytes. Our model is able to describe the local solvation state of the electrolyte, especially in highly concentrated electrochemical double layers.

To characterise the ion-solvent interaction, two parameters per ion species are chosen: The maximum number of solvent molecules binding to a single ion and the binding energy per bound solvent molecule. This form of the free energy yields plausible results in equilibrium condition, though we have not yet tested this free energy on a full battery model.

This work was funded by the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation) under Germany´s Excellence Strategy – EXC 2154 – Project number 390874152.

***Keywords****:* battery, modelling, electrolyte, solvation, electrochemical double layer.

1. Armand, M. ; Tarascon, J.-M. *Nature* **2008** 451, 652.
2. Park, J. et al *Frontiers in Chemistry* **2020** 8, 432. DOI: 10.3389/fchem.2020.00432
3. Lück, J. ; Latz, A. *Phys. Chem. Chem. Phys.*, **2018,** 20, 27804-27821. DOI: 10.1039/C8CP05113E
4. Dreyer, W. et al *Electrochem. Comm.* **2014**, 43, 75-78. DOI : 10.1016/j.elecom.2014.03.015
5. Schammer, M. et al *J. Electrochem. Soc.* **2021,** 168, 026511. DOI: 10.1149/1945-7111/abdddf
6. Schammer, M. et al *ArXiv.org* **2021** arXiv:2112.11511.