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Modeling the Voltage Dependence of Electrochemical Reactions at Solid-Solid and Solid-Liquid Interfaces in Batteries

KEVIN LEUNG, Sandia National Laboratories

Electrochemical reactions at electrode/electrolyte interfaces are critically dependent on the total electrochemical potential or voltage. In this presentation, we briefly review ab initio molecular dynamics (AIMD)-based estimate of voltages on graphite basal and edge planes [1], and then apply similar concepts to solid-solid interfaces relevant to lithium ion and Li-air batteries. Thin solid films on electrode surfaces, whether naturally occuring during power cycling (e.g., undesirable lithium carbonate on Li-air cathodes) or are artificially introduced, can undergo electrochemical reactions as the applied voltage varies. Here the onset of oxidation of lithium carbonate and other oxide thin films on model gold electrode surfaces is correlated with the electronic structure in the presence/absence of solvent molecules. Our predictions help determine whether oxidation first occurs at the electrode-thin film or electrolyte-thin film interface. Finally, we will critically compare the voltage estimate methodology used in the fuel cell community [2] with the lithium cohesive energy calibration method broadly applied in the battery community, and discuss why they may yield different predictions.

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[1] K. Leung and C.M. Tenney, J. Phys. Chem. C 117, 24024 (2013).
[2] J. Cheng and M. Sprik, Phys. Chem. CHem. Phys. 4, 11245 (2012).