Elucidating Interfacial Reactions in Rechargeable Alkali-Ion Batteries

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The reactions occurring at interfaces have a profound impact on the performance of a rechargeable alkali-ion batteries. In all-solid-state battery architectures, interfacial resistance – both in the grain boundaries of the solid electrolyte and between the solid electrolyte and the electrodes – are now the critical bottleneck limiting cell performance. We will present an exposition of how first principles approaches can help elucidate interfacial reactions and the impact of the resulting reaction products on cell performance.[1] We will demonstrate that by considering the cell at the limit of relatively fast/slow alkali diffusivity, thermodynamics approximations can be made that provide a useful picture of interfacial phase equilibria at a reasonable computational cost. A more accurate picture can be provided using kinetic approaches such as *ab initio* molecular dynamics, and sometimes, the predicted interfacial composition can differ substantially from those from thermodynamics approximations. I will highlight the importance of considering intercomponent interactions (e.g., electrode-electrolyte) as well as the properties of the interfacial products in designing the battery cell as a whole for optimal performance. Specific recommendations will include choice of cathode chemistry for a given electrolyte chemistry as well as selection criteria for buffer layers to mitigate interfacial reactions. Examples will be shown from both lithium-ion as well as sodium-ion battery chemistries.

References:

[1] Tang, H.; Deng, Z.; Lin, Z.; Wang, Z.; Chu, I.-H.; Chen, C.; Zhu, Z.; Zheng, C.; Ong, S. P. Probing Solid–Solid Interfacial Reactions in All-Solid-State Sodium-Ion Batteries with First-Principles Calculations. Chem. Mater. 2018, 30 (1), 163–173 DOI: 10.1021/acs.chemmater.7b04096.