Modeling of Interfaces in All-Solid-State Li-ion Batteries

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Paring solid electrolytes with Li metal anode and high voltage cathodes form promising all-solid-state Li-ion batteries with higher energy density and ultimately safety. With the rapid development of fast Li-ion conducting solid electrolytes, the major bottleneck for all-solid-state Li-ion batteries lies at the high interfacial resistance and Li dendrite growth. This talk will focus on new mechanism understanding obtained by atomisticly-informed multi-scale modeling approaches.

The high interfacial resistance is due to two main factors: physical contact and chemical effect. The chemical effect was captured by a new density functional theory (DFT) based model, which predicts the potential map inside a solid-state battery and determines the potential drop, electrostatic dipole, and space-charge layer at the electrode/solid-electrolyte interface.[1] This new physics insight unified the seemingly contradictory experimental observations and led to new device design rules to promote interfacial ion transport in future solid-state batteries.[2] The physical contact was described by combining contact mechanics and 1D Newman battery model. The model suggested how much pressures should be applied to recover the capacity drop due to contact area loss.

To simulate Li dendrite growth inside polycrystalline solid electrolytes, we coupled DFT calculations with the phase-field method. This model successfully explained the experimentally observed dendrite intergranular growth and revealed that the trapped electrons at grain boundaries and surfaces may produce isolated Li-metal nucleation, leading to a sudden increase of Li-dendrite penetration depth. Based on the model, we developed new dendrite resistant criteria by comparing the basic material properties for a number of solid electrolytes including LLZO, Li_3PS_4, LATP, and LiPON. [3][4]

These modeling advancements will be integrated into a new framework to guide the development of all-solid-state Li-ion batteries.

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