Modelling the Lithium Anode-Electrolyte Interface under Potentiostatic control

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ABSTRACT

The resurgence of the lithium anode in solid-state batteries has garnered considerable interest, primarily due to its standout qualities. Lithium metal anodes boast a high theoretical specific capacity of 3860 $mAhg^{-1}$and a low density of 0.53 gcm^{-3} at 25°C[1]. When paired with solid-state electrolytes, these characteristics hold the potential to revolutionise energy storage technology by enabling the development of compact, high-energy-density batteries capable of achieving output voltages up to 6V, all while eliminating the need for hazardous and flammable organic solvents.

In this study, we conduct a computational analysis to investigate the surface dynamics of the lithium anode, with a focus on how its low energy surfaces potentially adapt or reorganise when subjected to an applied voltage. By calculating the surface energies of said lithium surfaces under such conditions, we can construct Wulff shapes—a technique that visually elucidates how the anode's exposed surfaces may evolve under bias. This approach not only provides essential structural insights for future research on interfaces with solid-state electrolytes but also deepens our comprehension of key phenomena. These include surface reconstruction, dendrite formation, degradation mechanisms, and the role of electrolyte therein.

Our computational investigations are carried out under a consistent applied bias, leveraging the grand canonical[2] approach within ONETEP[3], a state-of-the-art, linear-scaling density functional theory programme.

References

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