First Principles Modeling of the Interface Between a Solid State Lithium Thiophosphate Electrolyte and a Lithium Metal Anode

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First principles modeling of the interface between a solid state lithium thiophosphate electrolyte and a lithium metal anode

N. A. W. Holzwarth and N. D. Lepley
Department of Physics, Wake Forest University
Winston-Salem, NC 27109, USA

Recently, there has been progress in improving the conductivity and stability of solid electrolytes such as Li_3PS_4.[1] We report the results of first principles computer modeling studies of ideal Li_3PS_4 electrolyte interfaces with metallic Li anodes.

For a variety of interface configurations, computer modeling studies show that Li_3PS_4 surfaces are structurally and chemically altered by the presence of Li metal. On the other hand, experiments have shown [1] that an electrochemical cell of Li/Li_3PS_4/Li can be cycled many times. One possible explanation of the apparent stability of the Li_3PS_4 electrolyte/Li metal interface, is that a stable thin buffer layer is formed during the first few cycles. In order to computationally explore this possibility, we modeled a "thin film" buffer layer of Li_2S on the surface of the electrolyte. Using first principles techniques described in previous work,[2] stable electrolyte-buffer layer configurations were found. Results for the idealized configurations indicate that a thin film of Li_2S can provide a protective buffer layer to stabilize the interface between the Li_3PS_4 electrolytes and Li metal anodes.

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