Modelling the Polymer Electrolyte/Lithium-Metal Interface by First Principle Calculations

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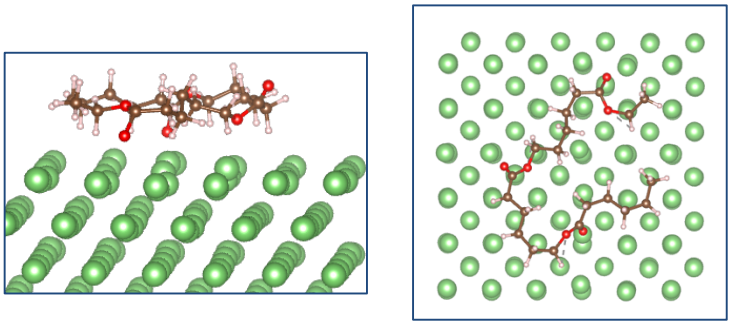
Lithium metal combines the lowest reduction potential in the electrochemical reactivity series with a high theoretical specific capacity, and using metallic Li as anode would therefore significantly improve the energy density of the Li-battery. There exist, however, some challenges in the application of the Li metal electrode, such as safety risks and low coulombic efficiency [1]. In recent years, there has been a growing interest to find more stable electrolytes when in contact with the reactive Li electrode in Li-metal batteries. It has in this context been found that solid polymer electrolytes (SPEs), formed by doping a polymer with a lithium salt, are promising candidates, which can provide both high mechanical stability and better battery safety [2,3]. The major disadvantage of SPEs – their low inherent ion conductivity – can be resolved by a somewhat higher operational temperature.
We have, in a number of studies [4-6], modelled the Li metal/electrolyte interface using different simulation techniques. In this current study, we apply computational materials modelling to investigate the interface between the ion-conductive polymeric systems and Li metal surfaces by first principle calculations. To this end, Density Functional Theory (DFT) have been used to study several potential SPE host polymers such as poly(trimethylene carbonate) (PTMC), poly(vinyl alcohol) (PVA) and polycaprolactone (PCL) (shown in Fig. 1), in order to get insights into their electronic structures and their stability when in contact with the Li metal surface. Using this knowledge, conclusions are drawn on which ion-conductive polymers are stable at the Li-metal surface, and which can adhere well to it.



(a) (b)

Fig. 1. (a) Side and (b) top views of a PCL chain adsorbed on the Li metal surface. Green, red, brown, and light pink spheres denote Li, O, C, and H atoms, respectively.

References:

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