

Li/C composites as anodes for high energy density rechargeable Li batteries

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Current lithium (Li) ion batteries consisting of graphite as the anode and intercalation materials (such as LiCoO_2 , $\text{LiNi}_x\text{Co}_y\text{Mn}_{1-x-y}\text{O}_2$, $\text{LiNi}_{0.8}\text{Co}_{0.1}\text{Al}_{0.1}\text{O}_2$) as the cathodes have almost reached their theoretical energy density of 300 Wh/kg. As a result, exploring high energy density batteries is urgent. Li-metal taking place of the graphite as the anode has several advantages. On one hand, it has a low operational voltage (-3.04 V versus standard hydrogen electrode) and a high specific capacity of 3860 mAh/g offering the battery with high energy density. On the other hand, when the Li-metal as the anode, the cathodes can be extended to Li-free or Li-deficient materials, which means the cathodes have more choices. For instance, S, O_2 , conversion reaction-type materials can be optional. While, the introduction of Li-metal anodes brings in several challenges^[1-3]. Firstly, Li-metal is highly electrochemical and electrochemical active to have undesirable reactions with conventional carbonate-based liquid electrolytes, leading a low Coulombic efficiency and large polarizations. Secondly, the formed Li dendrites due to uneven Li^+ distribution during plating and stripping has the chances of percolating separators leading to short circuits. Thirdly, Li metal is a host-free material ($\sim 5 \mu\text{m}$ have a capacity of 1 mAh/cm²), thus it suffers from a huge volume change, which brings in difficulty in the cell design.

Tremendous efforts have been paid to improve the electrochemical performance of the Li metal via surface coating a Li-ion conductor in the carbonate-based electrolyte system. Reported in *Advanced Energy Materials*, Jung-In Lee *et al.* applied an Li-ion-conductor Li_2TiO_3 on Li metal film to fabricate an artificial solid electrolyte interphase (SEI), which effectively improved its cycling performance^[4]. It is demonstrated that the Li-ion coating layer can suppress the Li dendrite formation and prevent side reactions between the lithium metal and liquid electrolyte. This work is meaningful. It would be more interesting to explore the stability and transport properties of the artificial SEI layer Li_2TiO_3 during stripping and plating as functions of current rate and capacity. Once the Li directly plates on the Li_2TiO_3 surface, the side reaction with the liquid electrolyte is inevitable.

A 3D framework to host Li metal in the solid state electrolytes seems to be an ideal option to solve the Li-metal issues mentioned above. Here we propose low-cost porous carbon with Li as Li/C composites for the anodes. As demonstrated in Fig. 1, the carbon has porosity to provide a host for Li. Meanwhile, carbon has high electronic conductivity, which can be

considered to be an equipotential body for a homogeneous Li^+ distribution. Choosing proper solid state electrolytes, Li metal can be inert with the electrolytes to prevent side reactions. Considering the application demand of this composite material, the capacity of Li should be estimated to properly design the porosity and thickness of the carbon. The method on how to prepare Li and C composites need be carefully designed. Moreover, the Li deposition into the carbon pore should be controlled via surface modification to be Li-philic nature^[5]. As the volume change is inevitable in the solid state battery, an introduction of *in-situ* growth solid state electrolyte is favorable^[6].

In summary, low-cost Li/C composites as the anodes have the potential to offer high energy density rechargeable Li batteries, such as Li-S, Li- O_2 , Li- $\text{LiNi}_{0.8}\text{Co}_{0.1}\text{Mn}_{0.1}\text{O}_2$ battery. Co-operated with solid state electrolytes and surface modification, they may conquer the shortcomings of high volume change, high chemical activity at the Li-metal and electrolyte interface as well as prevent the dendrite formation.

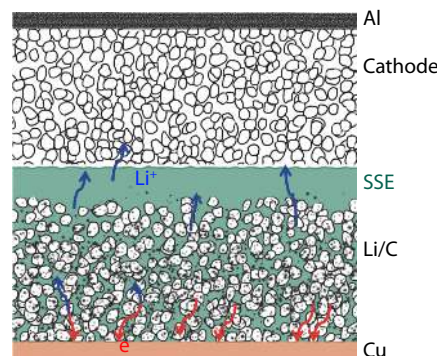


Fig. 1. (Color online) Li/C composites as ideal anodes in the Li batteries with solid state electrolytes (denoted SSE). Note that the cathode material is not discussed here and the cell is demonstrated at a discharge state.

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