

Remarkable improvement in cyclability of polymer lithium-ion batteries by a new electrolyte containing  
functionalized poly (methyl methacrylate)

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Lithium-ion batteries have a wide application range from powering small sized electronic devices to driving electrical vehicles due to their high specific energy and power density [1-2]. Such a high energy density is due to the large charge density of anode and cathode active materials. In spite of these advantages, a few challenges are still to be resolved for the future of lithium-ion batteries. One of the major issues is the safety concerns raised from volatility and flammability of conventional liquid electrolytes used in these batteries, which can severely limit their overall applications [3]. For the last two decades, consistent research has been focused on developing more reliable electrolytes for lithium-ion batteries. The most viable alternative to liquid electrolytes is a gel polymer electrolyte (GPE). Among all GPEs, PVdF-HFP is the most studied one because of its good electrochemical stability, plasticity and liquid electrolyte uptake [4-5]. However, this GPE suffers from performance fading after a few cycles due to the poor liquid electrolyte holding [5-6]. A wide variety of approaches have been studied to enhance the performance of PVdF-HFP based GPEs. Blending has been considered to be an effective method for modifying the GPEs with desired properties; however, achieving such an electrolyte often requires an addition of high amounts of another polymer, which adversely affects electrochemical properties of the PVdF-HFP matrix.

Presented herein is the fabrication of a new GPE for these batteries. This work is to overcome drawbacks of PVdF-HFP based GPEs while simultaneously maintaining their beneficial electrochemical properties such as high ionic conductivity at ambient temperature and excellent chemical stability. A functionalized poly(methyl methacrylate) (PMMA) with inorganic trimethoxysilane groups was synthesized by free radical polymerization and blended in low amounts with PVdF-HFP. The idea behind this work was to reduce the pore size, promote the homogeneity of its distribution and improve the stability of the GPE morphology by addition of a compatible polymer having inorganic domains into PVdF-HFP. Consequently, changes in the morphology of GPE due to its swelling after liquid electrolyte uptake could be prevented by means of the covalent bonding between the inorganic groups and polymer chains, which counterbalances the plasticizing effect of the electrolyte solution.

The resultant polymer blend displayed ionic conductivity as high as 4.7 mS/cm at room temperature and a wide electrochemical stability window of 4.8 V. Tested by coin type cells, the new GPE acts as a very effective electrolyte for lithium-ion batteries. The Li/GPE/LiCoO<sub>2</sub> cell delivered an initial discharge capacity of 140 mAh g<sup>-1</sup> based on the weight of LiCoO<sub>2</sub> active material. Discharge capacity of 131 mAh g<sup>-1</sup>, 94% of the initial value, and coulombic efficiency of almost 100% were obtained after one hundred cycles indicating very good liquid electrolyte upholding of the gel electrolyte during cycling of the battery cell.

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