Chemical Engineering Doctoral Defense

Electrode-coated Inorganic Separators for High Performance and Safe Lithium-Ion and Lithium-Metal Batteries

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Abstract

Lithium-ion and lithium-metal batteries are deemed to be the choice of energy storage media for the future. However, they are not entirely safe and their performance in terms of cycle life and charging rates is sub-optimal. A majority of these issues arise from the currently used flammable polyolefinic separators and carbonate solvent based electrolytes. This work utilizes in-house developed electrodecoated completely inorganic separators in combination with a fire-proof electrolyte to resolve the above stated concerns. Firstly, we compare the performance of the commercial polypropylene separator with an in house developed electrode coated quartz silica separator. The silica separator shows better performance in terms of electrolyte wettability, electrolyte uptake and better capacity retention ($\sim 15\%$) at high rates of discharge. It also improves the safety of the battery due to its high thermal stability. Subsequently, research on developing a completely safe lithium ion battery was conducted by replacing the traditional carbonate solvent based electrolyte with a fire-proof lithium bis-fluoro sulphonyl-imide salt/tri-methyl phosphate solvent electrolyte. A microporous in house synthesized silicalite electrode-coated separator is developed to functionalize the viscous fire-proof electrolyte and they are tested in a full-cell setup. The intra-particle pores of the silicalite separator result in a thinner and more robust solid electrolyte interface on graphite. This results in about 20~%higher capacity retention during long term cycling when compared to the polypropylene separator used in the same full cell. Furthermore, plate shaped y-alumina and silicalite electrode-coated separators with high tortuosity are developed and used in a lithium metal full cell battery, with the former separator having no intra-particle pores and the latter having them. The y-alumina separators show improvements in dendrite propagation prevention up to 3 C-rate of charge/discharge but a loss in active lithium was seen beyond the 75th cycle. However, microporous plate-shaped silicalite separators did not show any loss in active lithium even at 3 C-rate for 100 cycles due to the homogenized lithium ion flux at the anode, while also preventing dendrite propagation. Thus, high performance and safe, lithium ion and lithium metal batteries are enabled by utilizing specific property tuned inorganic electrode-coated separators.