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Mechanical robustness of composite electrode for lithium ion battery: Insight into entanglement & crystallinity of polymeric binder

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ABSTRACT

To investigate the correlation between the molecular weight of the polymeric binder in Li-ion battery electrodes and their adhesion properties, polyvinylidene fluoride (PVdF) with three different molecular weights of 500,000, 630,000, and 1,000,000 are selected for $LiCoO_2$ electrode fabrication. Using a surface and interfacial cutting analysis system, it is observed that, as the molecular weight of the PVdF increases, the adhesion strength not only in the electrode composite, but also at the electrode/current collector interface increases. This enhancement can be attributed to the increased polymeric chain entanglement and higher crystallinity of PVdF with higher molecular weight, which is confirmed using a microfluidic viscometer and differential scanning calorimeter, respectively. In summary, regardless of slightly higher electrode resistance, the $LiCoO_2$ electrode with a PVdF binder of high molecular weight shows better electrochemical performance during cycling test even at 60 °C due to its stable mechanical integrity.

1. Introduction

The energy density of the lithium-ion battery (LIB) has been steadily increased to fulfill the need of customers for longer battery life on their mobile electric devices as well as to enable their use in electric vehicles (EVs) with a mileage per charge comparable to internal combustion engines so that they can be used as vehicles in the long run [1,2]. In order to increase the energy density of LIBs, the electrode or cell design is as important as the development of new electrode materials with higher specific capacities; thus, without determining optimum design parameters, such as electrode composition, electrode loading level, and electrode density, among others, it will not be possible to maximize the energy density of LIB cells even with the use of new, improved electrode active materials [3–5]. Therefore, the role of inactive materials in LIB electrodes becomes more crucial with increasingly complex design levels. For example, carbon nanotubes have already started being mixed with conventional spherical conductive carbon for highly loaded LIB electrodes, although this causes new issues, such as slurry mixing or stability problems [6].

Aside from the abovementioned efforts to improve the LIBs, minimizing the total amount of the polymeric binder in LIBs is also attracting attention among researchers [7,8]. However, because polymeric binders are quite closely related to the maintenance of the mechanical integrity of the electrodes in LIBs, they should be handled carefully. In particular, in the case of large-format LIBs for EVs and energy storage systems (ESSs), currently, there is no alternative, but to use highly loaded electrodes even for long service times. Considering the significantly wider operating temperature range of −30 to 52 °C for EVs [9,10], decreasing the polymeric binder content to increase the energy density of LIBs might result in severe LIB reliability issues. Some previous works have reported on the delamination problems of the electrode layer from the current collector in LIBs attempting to explain the sudden capacity drop observed during high-temperature cycling [11–13]. However, there are no clear guidelines or reports in literature on optimum binder conditions for EV or ESS applications. In addition, till recently, no appropriate analysis tools were proposed or developed to understand the adhesion properties within electrodes as well as at the

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