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A crosslinked nonwoven separator based on an organosoluble polyimide for high-performance lithium-ion batteries

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ABSTRACT

Thermostable polymers offer excellent thermal and mechanical stabilities, and in this study, an organosoluble polyimide (PI) was employed to fabricate high-performance separators for use in advanced lithium-ion batteries (LIBs). The organosoluble PI was synthesized and its nanofibrous membrane was fabricated via electrospinning. A subsequent heat treatment induced thermal crosslinking between the PI nanofibers to improve the heat resistance of the separators. PI nanofibrous membrane exhibited excellent wettability toward the liquid electrolyte, resulting in a greatly improved rate capability and cycle life. These results suggest that organosoluble PI nanofibrous membranes have a high potential for use as separators in high-performance LIBs.

Introduction

The cell size and energy density of lithium-ion batteries (LIBs) are continuously increasing to meet the requirements of range-extended electric vehicles (EVs) or low-cost energy storage systems (ESSs) [1–3,17,24–26]. Therefore, more electrode-active materials are needed within a limited cell volume [20], and this can be accomplished by minimizing the amounts of the inactive components, such as the separator, current collector, or polymeric binder. As a result, LIB cells have become more dangerous because they are more likely to explosively release energy under abuse conditions; thus, we need to secure the safety of large-format and high-energy-density LIBs by using other strategies.

In most commercial LIBs, porous polyolefin separators, specifically polyethylene (PE) and polypropylene (PP) separators, are used to prevent physical contact between the positive and negative electrodes while permitting free ionic transport within the cell. However, polyolefin separators suffer from severe thermal shrinkage at high temperatures due to their inherent low melting points, and this critical drawback occasionally leads to battery explosions. To solve this problem, some thermostable polymers have been employed in the form of nanofibrous separating membranes that are fabricated via electrospinning [4,5,18,19,21]. Polyimide (PI) is a high-performance, engineered polymer that has been widely used in various advanced technologies due to its excellent thermal and mechanical stabilities [18,19,22,23,27–30]. However, PI is often insoluble in most organic solvents due to the rigid structure of its backbone chains, and thus, it has limited applicability.

In this study, organosoluble PI was prepared via a polycondensation reaction with bicyclo[2,2,2]oct-7-ene-2,3,5,6-tetracarboxylic dianhydride (BCDA) and 4,4-oxydianiline (ODA). Then, PI nanofibrous membranes were fabricated via electrospinning to obtain thermostable separators for the LIBs. Differential scanning calorimetry (DSC), thermogravimetric analysis (TGA) and thermal shrinkage tests were conducted to investigate the thermal stability of the PI nanofibrous membrane. Additionally, the affinity of the PI nanofibrous membrane for the liquid electrolyte was evaluated by examining the liquid electrolyte wettability and uptake amount. Finally, the electrochemical performance of the PI nanofibrous membrane was compared with that of a commercial PP separator by fabricating coin cells with $LiCoO_2$/graphite cell chemistry.

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