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Full Length Article

Electron beam induced strong organic/inorganic grafting for thermally stable lithium-ion battery separators

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ABSTRACT

A tailored interface between organic and inorganic materials is of great importance to maximize the synergistic effects from hybridization. Polyethylene separators over-coated with inorganic thin films are the state-of-the art technology for preparing various secondary batteries with high safety. Unfortunately, the organic/inorganic hybrid separators have the drawback of a non-ideal interface, thus causing poor thermal/dimensional stability. Here, we report a straightforward method to resolve the drawback of the non-ideal interface between vapor deposited SiO_2 and polyethylene separators, to produce a highly stable lithium-ion battery separator through strong chemical linking generated by direct electron beam irradiation. The simple treatment with an electron beam with an optimized dose generates thermally stable polymer separators, which may enhance battery safety under high-temperature conditions. Additionally, the newly formed Si–O–C or Si–CH_3 chemical bonding enhances electrolyte-separator compatibility and thus may provide a better environment for ionic transport between the cathode and anode, thereby leading to better charge/discharge behaviors.

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1. Introduction

With the rapidly growing industrial importance of high energy density in a limited space, for use in applications such as mobile electronic devices of various shapes and electric or hybrid-electric vehicles (EVs or HEVs) [1,2], emerging high energy density lithium-ion batteries with high operating potential and high capacity has been receiving increasing attention [3]. Various emerging cathode and anode materials with high specific/volumetric energy densities have been explored [4–7]. However, the main approach to enhancing the energy density of lithium-ion batteries in terms of the separators (porous membranes) is simply to decrease the film thickness of polyethylene (PE) or polypropylene (PP), because these porous polymer films are not directly related to specific/volumetric energy density. Inherently, the thermal and dimensional stability of the polymeric porous film also decreases dramatically with decreasing film thickness; this characteristic has led to serious concerns about the safety issues of lithium-ion batteries, such as possible explosion and burning under high temperature or hard internal shorting between electrodes [8].

From the viewpoint of the basic requirements of battery separators, such as uniform pore size, high tensile strength, and chemical stability [9,10], ultra-thin polymeric separators coated with inorganic insulating particles, such as SiO_2 and Al_2O_3, allow easy integration into highly packed batteries with high energy density without raising safety concerns [11]. However, the additional inorganic particle layer on the PE separator with a thickness of approximately 3–6 μm decreases the volumetric energy density of the cells [12–14]. To increase the volumetric energy density of lithium-ion batteries, a new integration method of inorganic material into the PE separator is highly desirable. To overcome the drawback of conventional inorganic insulating particles coating PE separators and to improve the dimensional stability of the PE separator, alternative thin-film deposition of an inorganic layer inside the pores of a PE separator from atomic layer deposition (ALD), chemical vapor deposition (CVD) and sputtering methods have previously been suggested [15–17]. Although gas or vapor-phase deposition allows for enhanced thermal and dimensional stability, it is not guaranteed that the inorganic layer will be completely anchored on the surface of the PE separator, thus potentially weakening the positive effects of the inorganic layer and inducing delamination of the inorganic layer from PE during long-term cycling.

The porous structure of commercially available PE separators with sub-micron-sized channels is suitable for introducing thin

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