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Novel cellulose aerogel coated on polypropylene separators as gel polymer electrolyte with high ionic conductivity for lithium-ion batteries

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ABSTRACT

The low ionic conductivity of polypropylene (PP) separators has stymied their use as polymer electrolytes in high-power lithium-ion batteries. To improve the ionic conductivity, we coated PP separators with cellulose aerogel based on hydroxyethyl cellulose (HEC), via ice segregation induced self-assembly (ISISA). The coating was characterized via scanning electron microscopy (SEM), attenuated total reflection–infrared spectra (ATR-IR), thermogravimetric analysis (TGA), and differential scanning calorimetry (DSC). In addition, the electrochemical performances of the separator were evaluated by using a cell consisting of the coated separator, lithium foil as the counter and reference electrodes, and $LiFePO_4$ as the cathode. The porous cellulose aerogel-coated separator exhibited superior dimensional stability, electrolyte uptake, and hence a higher ionic conductivity and better cycling performance, than its non-coated counterpart. Furthermore, the separator was coated without the use of toxic solvents, thereby rendering the preparation process cost effective and highly efficient.

1. Introduction

Lithium-ion batteries with high energy density and excellent cycle life have attracted considerable attention as one of the most promising energy storage systems and suitable power sources for electric vehicles, hybrid electric vehicles, plug-in hybrid electric vehicles, etc. [1–4]. Most conventional lithium-ion battery separators are fabricated from polyolefins, predominantly polyethylene (PE) or polypropylene (PP), which have high mechanical strength and exhibit high electrochemical stability [5–7]. However, electrolyte infiltration of these types of separators is difficult, owing to their low polarity and low surface energies; this leads to poor compatibility between the separators and the electrolyte, as well as electrodes in the batteries, thereby resulting in poor ionic conductivity [8,9]. A separator that can be easily wetted by the electrolyte and has high ionic conductivity is therefore highly desirable.

Several strategies have been used to endow hydrophilic properties to intrinsically hydrophobic separators. In some cases, the polyolefins are replaced with hydrophilic non-wovens [10–14] or polymer fibers [15]. Although better hydrophilic properties (compared to those achieved by using polyolefins) are obtained by using these separators, thermal shutdown that prevents thermal runaway of the batteries is not ensured. Hydrophilic properties have also been endowed by coating organic materials [16–18], inorganic materials [19,20] or organic-inorganic mix materials [21,22] onto the surface of polyolefin separators. The inorganic nanoparticle coatings must be < 10 μm-thick, however, in order to maintain the toughness and open porous structures of the separator. Moreover, in the case of organic coatings, toxic solvents retained from the preparation stage, may adversely affect the electrochemical performances of the separator [23]. In addition, the preparation of inorganic-organic composite coatings entails several steps and complex processes.

Cellulose has attracted significant attention as a hydrophilic modification coating material, owing to its polarity and high thermal stability. For example, Xiao et al. [24] reported that a composite poly(vinylidene fluoride) (PVDF) membrane coated by methyl cellulose (MC) exhibited a high electrolyte uptake (138.6 wt%) and had a high lithium ion transference number (0.47). However, the use of cellulose aerogel as a coating layer for Li-ion batteries has barely been investigated. Cellulose aerogel is highly porous and exhibits excellent water retention and hence,

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constitutes a suitable candidate coating for modifying polyolefin separators [25,26].

As such, we prepared porous hydroxyethyl cellulose (HEC) aerogel and used ice segregation induced self-assembly (ISISA) to coat this gel onto a commercialized PP separator. The coated PP separator exhibits both excellent electrolyte uptake and ionic conductivity. Moreover, the separator was coated without the use of toxic solvents, thereby rendering the preparation process environmentally friendly, cost effective, and highly efficient.

2. Experimental

2.1. Materials

A PP separator (Gelgard 2400, porosity: 39%, 25 μm) was used as the polymer matrix of coatings and a contrast sample. HEC (250–450 mPa s, 25 °C), tri (hydroxymethyl) aminomethane (Tris), and dopamine were purchased from Aladdin Industrial Co. (Shanghai, China). Paraffin liquid and Tween 80 were

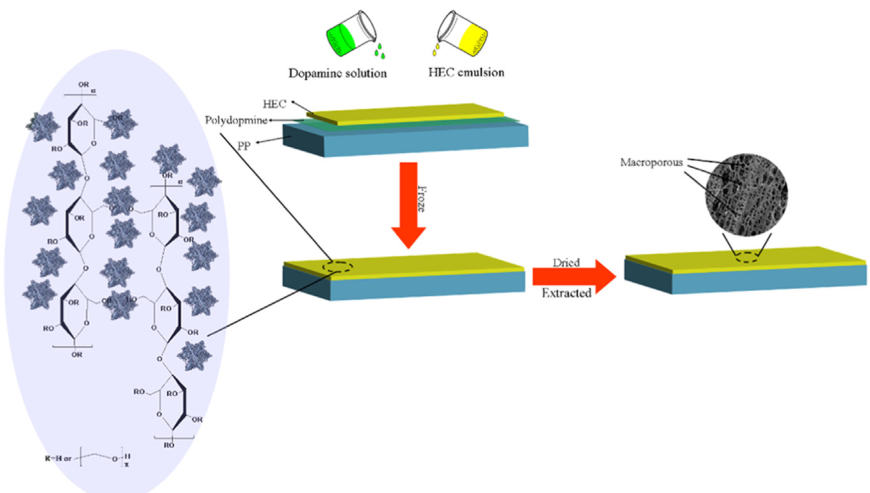

Fig. 1. The preparation of HEC aerogel coating for modification of the hydrophobic commercialized PP separator.

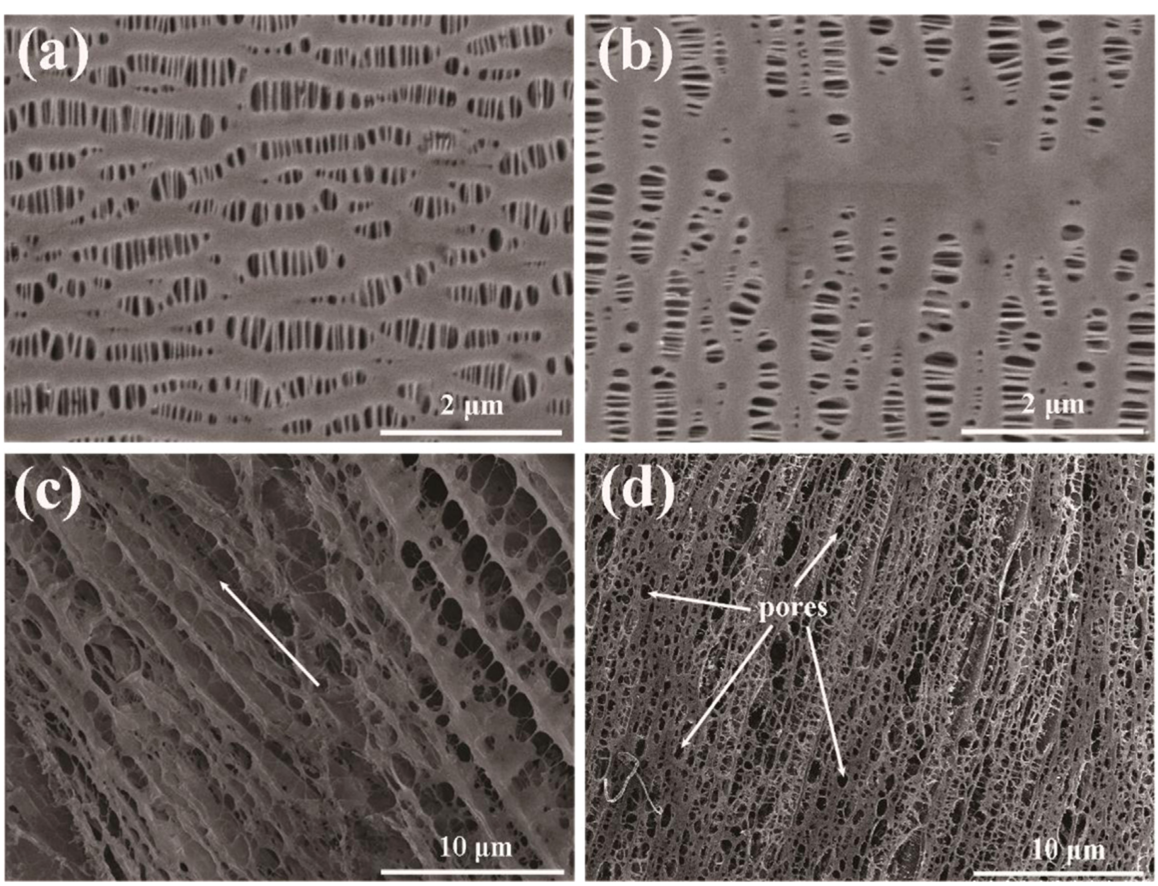

Fig. 2. The surface morphology of the PP and PP-HEC separators; the (a) PP separator and (b) dopamine-coated PP separator; the PP-HEC separator (c) before and (d) after extracting the paraffin liquid droplets.