



Correlation Studies Between Structural and Ionic Transport Properties of Lithium-Ion Hybrid Gel Polymer Electrolytes Based PMMA-PLA

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Abstract

In this work, an investigation on hybrid gel polymer electrolytes (HGPEs) comprising of polymethyl methacrylate (PMMA)-poly(lactic acid) (PLA) incorporated with lithium bis(trifluoromethanesulfonyl)imide (LiTFSI) was carried out. The HGPEs samples were characterized for their structural, thermal and ionic conduction properties via FTIR, XRD, DSC, and EIS. FTIR analysis showed the interaction between the PMMA-PLA hybrid polymer and LiTFSI through the appearance of peaks and peak shifts at the coordinating site on the polymer blend. The DSC analysis showed that the glass transition temperature (T_g) of HGPEs was decreased as the LiTFSI content increased, suggesting that the ion–dipole interaction decreased and led to the enhancement of the HGPEs system's amorphous phase. The ionic conductivity was calculated based on the Cole–Cole plot and the incorporation of 20 wt% LiTFSI into the hybrid polymer matrixes revealed that the maximum ionic conductivity was $1.02 \times 10^{-3} \text{ S cm}^{-1}$ at room temperature as the amorphous phase increased. The dissociation of ions and transport properties of the PMMA-PLA-LiTFSI systems was determined via the dielectric response approach and it was found that number density (η), mobility (μ), and diffusion coefficient (D) of mobile ions followed the ionic conductivity trend.

Keywords Electrical properties · Gel polymer electrolytes · Li^+ carrier · Ionic transport properties

Introduction

Over the past few decades, gel polymer electrolytes (GPEs) has been recognized as one of the promising technologies for applications in electrochemical devices, especially in lithium ion batteries. In GPEs, the liquid component is trapped in the polymer matrix, thus preventing leakage and improving the safety of the device application [1]. The presence of a liquid in GPEs also increases the cation mobility, thus leading to a boost of room temperature ionic conductivity [2]. The reported GPEs generally distribute high ionic conductivities

at a range of $\sim 10^{-4}$ to $10^{-3} \text{ S cm}^{-1}$ [3–5]. The implementation of GPEs also offers additional benefits, including being more mechanically and thermally stable, having sufficient electrochemical stability, and having the advantages of shape controllability [6,7]. Recent studies have identified several suitable polymer matrixes as hosts for GPEs. Among these polymers are poly(methyl methacrylate) (PMMA) [6,8], poly(ethylene oxide) (PEO) [9], polyacrylonitrile (PAN) [10], poly(vinylidene fluoride) (PVDF) [11,12] and polyvinyl chloride (PVC) [13].

Nevertheless, these single polymer-based GPE could not completely meet the requirements for practical applications in terms of long-term stability of cycle performance, superior mechanical and tensile strength, and good corrosion resistance [2,14,15]. To accomplish these desired properties, blending two different polymers is one method for enhancing the physicochemical properties of GPEs [12,16]. This method, also referred to as hybrid polymers in some cases, allows the polymers to interact chemically or physically, depending on their structural and physical properties against their parent components [17]. When two or more polymers are blended and achieve a homogenous phase, it leads to the formation of a miscible blend. In this

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