

Performance of PVDF-HFP-based gel polymer electrolytes with different pore forming agents

Wei Xiao · Xinhai Li · Zhixing Wang ·
Huajun Guo · Yan Li · Bo Yang

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Abstract Poly(vinylidene fluoride-*co*-hexafluoropropylene) (PVDF-HFP)-based gel polymer electrolytes (GPEs) with polyvinylpyrrolidone (PVP) and urea as a pore forming agent, respectively, were fabricated by phase inversion method. Physicochemical properties of the as-prepared polymer electrolytes were characterized by SEM, XRD, TG, electrochemical impedance spectroscopy (EIS) and linear sweep voltammetry (LSV). The results showed that the GPE membranes using urea as pore forming agent present a uniform surface with abundant interconnected micro-pores and possess up to 330 °C decomposition temperature; the XRD patterns indicate that adding urea into the polymer matrix can attain more amorphous areas than adding PVP; the reciprocal temperature dependence of ionic conductivity of as-prepared GPEs follows Vogel-Tamman-Fulcher relation and the ionic conductivity at room temperature is 2.212 mS cm⁻¹ for PVP pore forming GPEs and 2.823 mS cm⁻¹ for urea pore forming GPEs, respectively; the interfacial resistance of the Li/GPEs/Li cell using urea as pore forming agent can achieve a quick steady value of about 660 Ω cm⁻¹ lower than that of PVP of about 760 Ω cm⁻¹ during the same storage conditions; the electrochemical stability window of the GPEs with urea can be stable up to 5.0 V (vs. Li⁺/Li) at room temperature. The battery performance of the assembled Li/GPEs/Li-CoO₂ coin cell also showed that the cell using urea as pore forming agent in GPEs demonstrated excellent first charge/discharge rate and cycle performances. These excellent physicochemical and battery properties indicated that urea

can be used as a kind of excellent pore forming agent for polymer electrolytes in the lithium-ion polymer battery.

Keywords PVDF-HFP · Gel polymer electrolyte · Forming agent · Urea · PVP

Introduction

During the last decade, gel polymer electrolytes (GPEs) in the lithium-ion battery have received considerable attention for their excellent properties; for instance, they can render the systems lighter in weight, safer and more flexible in design and manufacture as compared with liquid electrolyte systems [1, 2]. GPEs are mainly made up of polymer matrix containing entrapped liquid electrolyte and suitable additives [1, 3, 4]. It is well known that some polymers and copolymers are successfully used as matrix material in lithium-ion battery, such as poly(methyl methacrylate) (PMMA), poly(oxethylene) (PEO), poly(acrylonitrile) (PAN), poly(vinylidene fluoride) (PVDF) and poly(vinylidene fluoride-hexafluoropropylene) (PVDF-HFP) [5–7]. Among them, PVDF-HFP is considered to be the most promising matrix as separator material, because it has relatively lower crystallinity due to the copolymerization effect between VDF and HFP compared with PVDF [3, 8]. The polymer electrolyte membranes used to entrap liquid electrolyte must possess abundant interconnected micro-pores. Therefore, some measures have been taken to improve the surface morphology of the polymer electrolyte membranes, such as addition of pore forming agents. Using poly(ethylene glycol) (PEG) as pore forming agent in the PEO-*co*-PMMA + SiO₂/lithium bis(oxalate)borate (LiBOB), composite polymer electrolyte system can undoubtedly enhance ionic conductivity and interfacial properties [9, 10],

W. Xiao · X. Li (✉) · Z. Wang · H. Guo · Y. Li · B. Yang
School of Metallurgical Science and Engineering, Central South
University, 410083 Changsha, People's Republic of China
e-mail: xwylyq2006@126.com