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Blended cathode materials for all-solid-state Li-ion batteries

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ABSTRACT

In this article, we report the effects of blended cathode materials in all solid-state lithium-ion batteries (ASLBs) with oxide-based inorganic-organic-hybrid electrolytes. A high capacity Ni-rich cathode material was used for high energy density in an ASLB, and $LiFePO_4$, with its robust olivine structure, was added to compensate for the degradation of the Ni-rich cathode material during cycling. In case of the blended cathode, the shortcomings of the parent material can be minimized by blending two cathode materials, and the blending ratio can be tailored to produce stable high energy and power densities. For these reasons, the structure, cycling stability, and rate performance of the blended $LiNi_{0.7}Co_{0.15}Mn_{0.15}O_2$/$LiFePO_4$ cathode material, for use in ASLBs with oxide-based inorganic–organic-hybrid electrolytes, was investigated by powder X-ray diffraction, field-emission scanning electron microscopy, Brunauer–Emmett–Teller sorption experiments, electrochemical impedance spectroscopy, and galvanostatic testing.

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1. Introduction

Currently, rechargeable lithium-ion batteries (LiBs) are widely used as power sources for xEVs, such as hybrid and plug-in-hybrid electric vehicles. Indeed, the recent increase in demand for electric vehicles has accelerated the development of new lithium batteries with stable and reliable performance [1–4]. Safety concerns surrounding LiBs remain major challenges due to the use of flammable organic liquid electrolytes. In this regard, all-solid-state Li-ion batteries (ASLBs) are promising candidates that may solve this problem because they use non-flammable inorganic solid materials as electrolytes and separators.

In particular, bulk-type ASLBs composed of composite electrodes and electrolytes that are based on the wet-slurry process have many advantages, including fabrication processes that are similar to those of commercialized LiBs, and high energy densities due to the ability to manufacture thick electrodes. The solid electrolyte (SE) material is the most important component of an ASLB, and its high ionic conductivity and mechanical properties are critical factors that determine the selection of a suitable SE [5]. In particular, solid polymer electrolytes (SPEs) based on a variety of polymers, such as poly(ethylene oxide) (PEO) [6,7], poly(vinylidene fluoride) (PVDF) [8,9], poly(vinylidene fluoride–hexafluoropropylene) (PVDF-HFP) [10,11], and polyacrylonitrile (PAN) [12], have been widely used because of their high flexibilities and low weights. However, in spite of these advantages, the low ionic conductivities ($<10^{-5}$ S cm^{-1} at room temperature), weak mechanical strengths, and poor stability windows of SPEs remain intrinsic challenges [13].

Due to the unsatisfactory properties of bulk-type ASLBs, inorganic–organic-hybrid electrolytes have recently been intensively investigated, especially composite membranes composed of inorganic materials such as garnet-type $Li_7La_3Zr_2O_{12}$ (LLZO), NASICON-type $Li_{1+x}Al_xTi_{2-x}(PO_4)_3$ (LATP), and polymeric Li matrices [14]. In particular, inorganic nanoparticles, such as LATP, play important roles as ionic conducting fillers in PEO-based composite electrolytes that enhance electrochemical properties and thermal stabilities, and protect the lithium dendrite. The PEO-Li salt matrix also reduces interfacial resistance due to its fast Li-ion conduction pathways, resulting in improved ASLB electrochemical performance [14,15]. In addition, in terms of compatibility with conventional electrode manufacturing processes, inorganic–organic-hybrid electrolytes are prepared in a comparable manner to current LiBs J. H. Choi et al. reported on the free-standing inorganic–organic-hybrid composite membrane prepared by similar way

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