



Contents lists available at ScienceDirect

Journal of Industrial and Engineering Chemistry

journal homepage: www.elsevier.com/locate/jiec

Effect of nanoparticles in cathode materials for flexible Li-ion batteries

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ARTICLE INFO

Article history:

Received 19 June 2019

Received in revised form 2 September 2019

Accepted 11 September 2019

Available online 28 September 2019

Keywords:

Li-ion batteries

Flexible lithium ion battery

Nanoparticle

LiMn₂O₄ cathode material

ABSTRACT

In this article, we report the effect of using nanoparticles for LiMn₂O₄ cathode materials in flexible batteries with organic–inorganic hybrid electrolytes. LiMn₂O₄ nanoparticles for the cathode are prepared by pyro-synthesis. Electrochemical measurement indicated the discharge capacities were 118.41 and 138.12 mAh g^{−1} and the coulombic efficiencies were 91.50% and 97.28% for the micro- and nano-LMO materials, respectively. This is attributed to the nano-LiMn₂O₄ material having particle sizes in the nanoscale dimension, and shorter diffusion paths combined with a large contact area at the electrode/electrolyte interface. Furthermore, the pouch-type cells demonstrated similar properties, with initial discharge capacities of 85.63 and 99.96 mAh g^{−1} and coulomb efficiencies of 79.27% and 90.27% for the micro- and nano-LMO cells, respectively. Nanoparticles allow Li⁺ ions to de-intercalate and intercalate very easily because of the very short lithium diffusion distance.

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Introduction

Market requirements for flexible and wearable devices have gradually increased for various applications such as smart watches, smart cards, healthcare, smart packaging, and wearable military and medical applications. The most important part of flexible electronic devices is the battery. Flexible rechargeable lithium ion batteries (LiBs) in particular have attracted significant attention because of their high flexibility, high energy density, superior rate performance, stable cycle life, and easy fabrication [1–5].

Flexible LiBs can be divided into three groups: one-dimensional (1D), two-dimensional (2D), and three-dimensional (3D) [6–15,39]. A representative 1D flexible LiB is a wire-type (cable type) battery that was reported by Kwon et al., in which the LiB can be deformed and twisted into any shape [15]. However, the use of these cable-type batteries in flexible electronic devices can be limited because of low energy density caused by the thin electrode layers required for minimizing degradation by mechanical stress. For 2D flexible batteries, paper-based LiBs have been intensively investigated because the porous and rough surface structure of the paper is adjustable for ion transportation, and its flexibility and bendable properties can result in ideal flexible batteries. The Cui

group has reported on the use of coated paper with single-walled carbon nanotubes (SWNTs) for improved electric conductivity. In addition, carbon nanotube (CNT)-based flexible 2D LiBs have been extensively developed because of their high bendability and high conductivity [14]. Finally, Chen et al. have reported on the formation of 3D flexible batteries using a 3D hydrogel electrode composed of CNTs and the conductive polymer PEDOT:PSS [12]. PEDOT:PSS is a dispersant for CNTs and active materials, and has a large role in forming a uniform conductive network, thereby securing mechanical flexibility. These batteries have the highest surface area capacity yet reported for flexible electrodes.

Conventional lithium secondary batteries are vulnerable to exploding due to leaks or short circuits because a liquid electrolyte and a polyolefin-based separator are used. Therefore, it is crucial to solve such a safety problem when manufacturing flexible batteries which require the ability to change shape. Polymer electrolytes have attracted attention as a solution to this problem, and extensive research has been conducted on several polymer electrolytes such as poly(ethylene oxide) (PEO), poly(acrylonitrile) (PAN), poly(methyl methacrylate) (PMMA), poly(vinyl chloride) (PVC), poly(vinylidene fluoride) (PVdF), and poly(vinylidene fluoride- hexafluoropropylene) (PVdF-HFP) [16–22]. Among the various polymer electrolytes, PVdF-HFP-based electrolytes have attracted significant attention due to the inherent properties of the polymer. This polymer is composed of both amorphous and crystalline phases, in which the crystalline PVdF phase acts to

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