Journal of Power Sources 332 (2016) 406–412

Contents lists available at ScienceDirect

Journal of Power Sources

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

Stretchable spiral thin-film battery capable of out-of-plane deformation

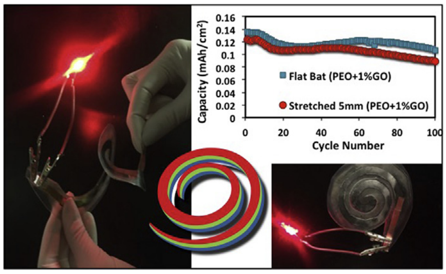
Mejdi Kammoun [a], Sean Berg [b], Haleh Ardebili [a], [b], *

[a] Department of Mechanical Engineering, University of Houston, Houston, TX 77204-4006, USA
[b] Materials Science and Engineering Program, University of Houston, Houston, TX 77204-4006, USA

HIGHLIGHTS

- Design and fabrication of spiral thin-film lithium ion battery is demonstrated.
- The stretchable battery is capable of large out-of-plane deformation up to 1300%.
- The battery exhibits an average capacity of 0.105 mAh/cm^2.
- The energy density of the battery stretched at 5 mm is 4.862 mWh/cm^3.
- The battery shows about 88% voltage retention after 9000 stretching cycles.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 23 May 2016
Received in revised form
8 September 2016
Accepted 16 September 2016

Keywords:
Lithium ion battery
Solid polymer electrolyte
Stretchable
Spiral

ABSTRACT

There is a compelling need for innovative design concepts in energy storage devices such as flexible and stretchable batteries that can simultaneously provide electrochemical and mechanical functions to accommodate nonconventional applications including wearable and implantable devices. In this study, we report on the design and fabrication of a stretchable spiral thin-film lithium ion battery that is capable of large out-of-plane deformation of 1300% while exhibiting simultaneous electrochemical functionality. The spiral battery is fabricated using a flexible solid polymer nanocomposite electrolyte film that offers enhanced safety and stability compared to the conventional organic liquid-based electrolyte. The spiral lithium ion battery exhibits robust mechanical stretchability over 9000 stretching cycles and an energy density of 4.862 mWh/cm^3 at ~650% out-of-plane deformation. Finite element analysis of the spiral battery offers insights about the nature of stresses and strains during battery stretching.

1. Introduction

Over the recent years, there has been an increasing demand for deformable energy storage devices including flexible lithium ion batteries (LIBs) with various sizes, shapes, and mechanical properties to integrate with bendable, implantable, and wearable devices and applications [1–4]. The development of flexible and stretchable batteries mainly involves the design and fabrication of mechanically compliant and reliable active and passive materials through a rational assembly process [5]. The main challenge in the fabrication of the stretchable LIBs is to achieve mechanical

---

* Corresponding author. Department of Mechanical Engineering, University of Houston, Houston, TX 77204-4006, USA.
E-mail address: hardebili@uh.edu (H. Ardebili).

http://dx.doi.org/10.1016/j.jpowsour.2016.09.093
0378-7753/

deformation while maintaining the high electrochemical performance of conventional LIBs including high capacity and cycling stability [6].

In conventional spiral-wound electrode batteries [7–10], the electrode area is typically aligned parallel to the z-axis of the battery, resulting in an efficient increase of the active surface area within a limited size rigid battery case. These rigid batteries often incorporate organic liquid or gel electrolytes. Conventional batteries composed of flammable organic liquid electrolytes are generally incompatible with flexible applications due to their rigid packaging, susceptibility to leakage and safety hazards that can lead to fires and explosions. The safety issue in conventional batteries can be mitigated through the replacement of the liquid with solid electrolyte made of glass/ceramic or solid polymer. However, the application of glass/ceramic electrolytes in flexible and stretchable batteries is limited due to their mechanical stiffness, therefore, deeming polymer-based electrolytes as the most suitable candidates for stretchable energy storage devices [1,6,11,12].

Several thin-film designs [2,13–22] and novel materials [11,23–30] have been developed to achieve deformability and flexibility in energy storage devices. A review of the literature on flexible and stretchable batteries has been extensively provided in previous work [1,2,12]. Rogers and co-workers [17] reported a stretchable Li ion battery comprised of segmented active materials connected through stretchable serpentine electrical interconnects. The battery is composed of $Li_4Ti_5O_{12}$ and $LiCoO_2$ electrodes and liquid/gel electrolyte, and exhibits an areal capacity of 1.1 mAh cm^{-2} at C/2 and 300% strain capability. Another study by Kwon et al. [20] reported the design of a flexible cable-type Li-ion battery based on a hollow multi-helix Ni–Sn anode, a modified polyethylene terephthalate separator, and a $LiCoO_2$ cathode coated onto an aluminum wire. The electrolyte used in this battery was a liquid organic electrolyte injected at the center of the electrode assembly. The cable battery exhibited a stable reversible capacity of 1 mAh cm^{-2} with discharging rate of 0.1C between 2.5 and 4.2 V until 10 cycles, and demonstrated high mechanical flexibility and strength under severe bending and twisting conditions.

In the present work, a spiral design is proposed for a stretchable thin-film Li-ion battery (<1 mm thick) that is capable of significant out-of-plane deformation of 1300% while simultaneously providing electrochemical functionality (Fig. 1). The stretchable spiral battery is fabricated based on solid polymer nanocomposite electrolyte composed of polyethylene oxide (PEO) and 1 wt% graphene oxide (GO) and is encapsulated with plastic lamination sheet. The solid PEO/GO electrolyte offers higher ionic conductivity and transference number relative to the pure polymer electrolyte. Cyclic voltammetry of the spiral battery confirms that the PEO/GO electrolyte is compatible with the conventional battery electrodes (i.e. lithium cobalt oxide and graphite) and no side reactions are observed during the polarization. The spiral Li-ion battery displays robust mechanical stretchability over 9000 stretching cycles and an energy density of 4.862 mWh/cm^3 at 650% out-of-plane deformation and provides an average capacity above 0.1 mAh/cm^2 in different stretching configurations. The finite element analysis of the spiral battery provides insights on the torsional stresses and displacements in the battery during stretching. Spiral batteries based on solid polymer electrolyte offer enhanced safety and can be utilized in a wide range of applications, including deformable or irregularly shaped medical implants, piezoelectric technologies, robotic applications and textile industries.

2. Experimental

2.1. Polymer electrolyte fabrication and characterization

Polyethylene oxide (PEO) with chemical formula C_{2n+2} H_{4n+6} O_{n+1}, and 100,000 Mw was purchased from Sigma Aldrich. Lithium perchlorate salt ($LiClO_4$) was purchased from Sigma Aldrich with 99.99% trace metals basis and stored inside the dry glove box under constant Argon flow (H_2O < 0.5 ppm). Solid polymer electrolyte films were prepared by solution casting method. The electrolyte solution was prepared by mixing 2 g of PEO and 0.3 g of Li salt (ether-oxygen-to Lithium ratio (EO/Li) of 16:1) in a 4 oz. jar half filled with the solvent acetonitrile (C_2H_3N). 1 wt% of Graphene Oxide (GO) content was added to the solution to prepare the nanocomposite film. The nanoscale GO powder was purchased from Graphene Supermarket and synthesized using the Hummer's method (Supporting Information). After sonication for 30 min in Branson 3510 Sonicator, the resulting viscous solution was then poured into a Teflon petri dish (area = 76.9 cm^2) and dried at room temperature for ~24 h, to obtain free-standing solid electrolyte films with a thickness of ~200 μm. To ensure the removal of the solvent, the membranes were vacuum dried at 50 °C for an additional 24 h and stored in the glove box prior to testing.

Thermogravimetric analysis (TGA) data of the polymer nanocomposite electrolyte were collected using TA Instruments Model Q50 TGA under nitrogen atmosphere, from the room temperature to 500 °C, at a scan rate of 20 °C/min. Flammability tests of both

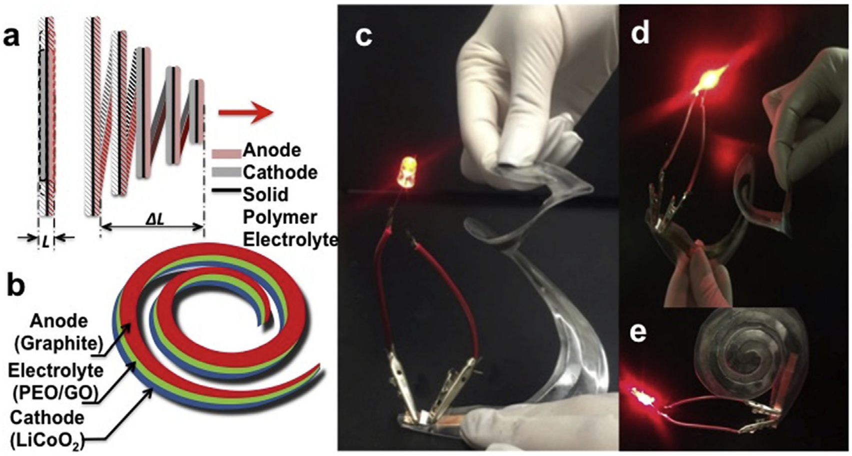


Fig. 1. (a)–(b) The schematics of the spiral Li-ion battery, (c)–(d) photo image of the fabricated spiral battery in stretched positions lighting a red LED, (e) spiral battery in flat position. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)