



# Ultra-thin flexible screen printed rechargeable polymer battery for wearable electronic applications



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## ABSTRACT

This research has demonstrated how an ultra-thin rechargeable battery technology has been fabricated using screen printing technology. The screen printing process enabled the sequential deposition of current collector, electrode and separator/electrolyte materials onto a polyethylene terephthalate (PET) substrate in order to form both flexible and rechargeable electrodes for a battery application. The anode and cathode fabricated were based on the conducting poly (3,4-ethylenedioxythiophen): poly (styrene sulfonate) (PEDOT: PSS) and polyethyleneimine (PEI) which were combined to form the electrodes. The difference in the oxidation level between the two electrodes produced an open circuit voltage of 0.60 V and displayed a practical specific capacity of 5.5 mAh g<sup>-1</sup>. The battery developed had an active surface area of 400 mm<sup>2</sup> and a device thickness of 440 μm. The chemistry developed during this study displayed long-term cycling potential and proves the stability of the cells for continued usage. This technology has direct uses in future personal wearable electronic devices.

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## 1. Introduction

There is a wide range of wearable electronic applications which require direct energy supplies to allow for self-contained operations. These applications demand a combination of; very low cost fabrication, low volume, light weight, flexibility, environmental friendliness and robust performance. Typical examples of power needs for such optimised components are 10 μW for a low power RFID chip, 17 μW at 1.4–1.6 V for an operational amplifier, 20 μW at 0.9 V for a GMR-switch or 5 μW cm<sup>-2</sup> for the update of an e-Ink display [1].

Printed batteries are reported to have lower durability, failure due to defects and non-uniform surfaces, attributed to difficulty in obtaining optimum materials and their associated challenges for formulation into an ink [2]. However, a completely printable battery is attractive because it gives freedom in form for integration with wearable devices that can only be commercially delivered through the use of printing technology. Energy source requirements for wireless sensor node applications for primary and rechargeable printable battery are low cost, high safety and small size [3]. However, despite these possibilities, the applications

of such thin-film batteries for different products are currently limited by their cost of production [1].

With the miniaturisation of electronics in mind, the design of batteries has changed over the past decades, and different types of prismatic or other thin batteries have been developed [3]. Several attempts have been made to produce printed batteries commercially. Thin primary batteries using the zinc–manganese chemistry have been manufactured by means of printing and are currently in use [4–8]. These technologies are still at an early stage, and the ability to print these will create new avenues for manufacturing novel miniaturized devices. Large area rechargeable printable battery technology, as would be required for wearable technology is still in its early phase of development.

The physical and electrical contact within a flat thin film battery cell as well as working duration, have been reported as limitations in design. These may be addressed through the design of anode and cathode components, through sealing film layers, or by adding binders or adhesives to the various components of the system [9]. As an example, an inkjet printed zinc–silver 3D battery has been reported having an energy density of 3.95 mWh cm<sup>-2</sup> [10]. This system is formed by electroplating zinc from a ZnO solution, but is limited by the challenge of dissolving ZnO in the alkaline electrolyte. In order to avoid this issue, there has been the development of an alkaline gel electrolyte to form a planar printed

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primary 2D silver–zinc battery to obtain an area energy density of  $4.1 \pm 0.3 \text{ mWh cm}^{-2}$  [11]. A recently developed flexible zinc–air battery has been produced by screen-printing a zinc/carbon/polymer composite anode, and a vapour polymerised PEDOT cathode onto two sides of a photo quality paper [12]. The lithium chloride and lithium hydroxide electrolyte was inkjet printed in eight layers to be absorbed within the papers cross-section and therefore, between the two electrodes [12].

Substantial work has been undertaken to develop secondary thin-film batteries. For example, a thin-film Li-ion microbattery has been developed using sputtered electrodes on a glass surface and a micro-injected sol–gel electrolyte [13]. A screen printed nickel metal hydride (Ni-MH) rechargeable battery on thin, flexible roll-fed plastic materials has been fabricated and a capacity of 32 mAh was reported [5]. A flexographically printed rechargeable Zn–MnO<sub>2</sub> battery with MnO<sub>2</sub> cathode, zinc anode and ionic gel electrolyte consisting of a 1:1 mixture of poly(vinylidene fluoride-co-hexafluoropropene) (PVDF-HFP) and 0.5 M solution of zinc trifluoromethanesulfonate (Zn + Tf<sup>-</sup>) salt dissolved in 1-butyl-3-methylimidazolium trifluoromethanesulfonate (BMIM + Tf<sup>-</sup>) ionic liquid has been developed [14]. This yielded a first cycle capacity of  $0.13 \text{ mAh cm}^{-2}$  with a 72% drop in capacity for successive cycles, with a reported reversible capacity of  $0.05 \text{ mAh cm}^{-2}$  [14]. The decrease in capacity demonstrates a significant loss of active material after a few cycles, thereby reducing its use in practical applications. Some of the reported disadvantages of the Zn–MnO<sub>2</sub> batteries are rapidly decreasing capacity with cycling and depth of discharge, limited cycle life and higher internal resistances when compared with NiMH [15].

A flexible Ag–Zn rechargeable battery has been developed using screen-printing on tattoo paper. This uses an alkaline gel electrolyte [16] and has the advantage of being thin, light weight, flexible, inexpensive and biocompatible. This configuration provided a maximum discharge capacity of  $2.1 \text{ mAh cm}^{-2}$ , but exhibited limited cycling capability and capacity loss due to high internal resistance [16]. Various other configurations of batteries include microfabricated alkaline Ni–Zn [17] and laser direct-writing of Zn–AgO micro-batteries [18], Li-ion electrode microarrays [13] and thin-film lithium batteries have been developed by several groups in recent years [4] and [19–22], but none of these have been printed on flexible substrates.

Based on the preceding discussion the limitations of the thin-film technologies can be broadly summarized as having lower capacities, recharge cycle limitations, being not fully printable and exhibiting challenges relating to their assembly and temporal stability. This leads to an interest in developing an all polymer based battery technology. In previous work, a battery concept comprising PEDOT: PSS and PEDOT: PSS with PEI electrodes covered with a PSSNa electrolyte demonstrated low open circuit voltage (OCV) of 0.5 V for a co-planar polymer battery fabricated using casting [23]. In this configuration, the two electrodes are deposited by casting, leaving a gap between them and the electrolyte was then deposited over this gap again by casting. This design led to an uneven current density distribution across the electrodes, resulting in increased internal resistance and lower performance.

In this work, we use the concept set out in [23] as a starting point. However we differentiate through the development of a layered architecture in which electrodes that have a large area in contact with the electrolyte are fabricated by screen printing and a separator that can also be fabricated by screen printing is placed between the electrodes to form the battery. The separator has to serve two purposes, the first is to facilitate ionic conduction while at the same time providing electrical insulation. This will be referred to as an electrolyte–separator in the following text.

The strategic aim of this design is to increase battery capacity and to use a fabrication route that will facilitate large scale manufacturing by printing. In a stacked configuration (Fig. 1), the electrodes, electrolyte and an insulator/separator are fabricated as a series layers. The stacked design is more suited for high duty applications due to a better energy density. The very short, vertical ion path through the consistent thickness electrolyte–separator layer to the electrode layers leads to much higher charge and discharge currents and uniform performance across the electrodes. Therefore, the shape or configuration of the cell influences the battery capacity as it affects factors such as internal resistance due to ion mobility and heat dissipation [15]. To prevent short circuits between the electrodes, a pinhole-free separator now becomes very important. A major manufacturing problem is the handling of the electrolyte/separator and a seal against water vapour is necessary in order to avoid dehydration and complete breakdown of the battery [5].

Presently, there are major hurdles in formulating such a printable electrolyte: giving a formula that remains wet and stable over the battery life. Currently, no printable solution has been formulated, which prevents production of batteries on an industrial scale. With regard to performance, within this work we also demonstrate for the first time the cyclic charge–discharge behavior of this battery design.

Thus in this research work, we address have overcome some of the challenges plaguing the area of flexible printable wearable batteries to demonstrate an approach that facilitates integration into a roll-to-roll (R2R) process. Once integrated into a R2R process, thin-film batteries have the potential to become one of the next generation of power sources for portable electronic applications due to the unique possibility for low cost, high-volume production on flexible substrates.

## 2. Experimental methods

### 2.1. Materials and reagents

A 175  $\mu\text{m}$  thick heat stabilized SU320 polyester film from HiFi Industrial films was used due to its low surface roughness and ability to withstand temperatures up to 150 °C without deformation. This transparent film also allows visual examination of the printed layers for pinholes.

Silver polymer ink (C2080415P2) and carbon ink (C2050503P1) were purchased from Gwent Electronic Materials (GEM, Gwent, UK). The anode and the cathode comprise silver and carbon layers. The silver ink has been chosen to provide a high level of conductivity, as this is used as a current collector and the highly conductive carbon ink chosen provides better adhesion to the underlying silver layer in an attempt to maintain lower internal resistance of the battery and to provide protection for the silver layer from undergoing redox reactions. The PEDOT: PSS (EL-P-5015) ink was purchased from Agfa. PEDOT: PSS is one of the most explored organic conducting polymers in electrochemical devices due to its higher conductivity and stability [23]. It is regarded as a promising material for electronic organic devices as it enables the fabrication of cost-effective, flexible devices through mass production [23–24].

Therefore, both the anode and the cathode were made of PEDOT: PSS. Additionally, on the anode, a polyethyleneimine (PEI) layer was applied in order retains the air stability of the electrode and the PEDOT: PSS layer underneath [23]. PEI, xanthan gum and hydroxypropyl cellulose (HPC) were obtained from Sigma–Aldrich. Using polymer electrolytes instead of aqueous electrolytes in batteries reduces complications significantly as it simplifies encapsulation and hence increases shelf life and the range of

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