Edited by

Senentxu Lanceros-Méndez | Carlos Miguel Costa

PRINTED BATTERIES

Materials, Technologies and Applications

WILEY

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Preface & Acknowledgements

He who sees things grow from the beginning
will have the best view of them.

Aristotle (384 BC–c. 322 BC)

Printed batteries are an excellent alternative to conventional batteries for an increasing number of applications such as radio frequency sensing, interactive packaging, medical devices, sensors, and related consumer products. These batteries result from the combination of conventional battery technologies and printing technologies. Printed batteries are increasingly being explored for highly innovative energy storage systems, offering the possibility for better integration into devices and novel application areas.

In this context, the main motivation of the present book is to offer the first comprehensive account on this interesting and growing research field providing the main definitions, the present state of the art, the main research issues and challenges, and the main application areas. In this scope, this book summarizes the frontline research in this fascinating field of study, presented by selected authors with truly innovative and preponderant work.

The book provides an introduction to printed batteries and the current state of the art on the different types and materials, as well as the printing techniques for these batteries. Further, the main applications that are being developed for those printed batteries are addressed as well as the principal advantages and remaining challenges in this research field.

The first chapter provides a general overview of the area of printed batteries. It deals with definitions and the main printed batteries types such as lithium-ion, Zn/MnO_2 and related systems. The advantages and disadvantages of printed batteries are discussed and the main applications summarized. Chapter 2 describes the printing techniques used for the production of printed batteries and gives a brief description of materials, substrates and the process chain used in printed batteries. Chapter 3 deals with the important issue of the influence of slurry rheology on electrode processing through its formulation, preparation technique, coating and drying systems. Moreover, the rheological characteristics of the electrode slurry are described.

Chapter 4 focuses on the polymer electrolytes used for the development of printed batteries. The state of the art on polymer electrolytes produced with different printing techniques is described in this chapter, as well as the electrolytes used in conventional and lithium-ion batteries.

The subject of Chapter 5 is the design of printed battery components. This chapter focuses on printed material layers for the electrodes used in Zn/MnO_2 batteries, lithium-ion batteries, and related systems.

Chapter 6 presents the main applications of printed batteries. Power electronics, RFID, sensors and actuators, medical and energy-harvesting devices are presented and discussed.

Taking into account the different applications of printed batteries, Chapter 7 provides an industrial perspective on printed batteries considering relevant industrial aspects such as layout considerations, current collectors, carrier substrates and multifunctional substrates, among other topics.

Finally, Chapter 8 summarizes some of the main open questions and challenges and the outlook for this research field.

This book would have not been possible without the dedicated and insightful work of the authors of the different chapters. The editors truly thank them for agreeing to devote their precious time to this enterprise. We thank them for their kindness, dedication and excellence in providing high-quality chapters illustrating the main features, challenges and potential of the area of printed batteries. It has been a pleasure and an honor to work with you in this important landmark in the field!

Additionally, this book would not have been possible without the continuous dedication, support and understanding of our research group colleagues both at the Center of Physics, University of Minho, Portugal, and the BCMaterials, Basque Center for Materials, Applications and Nanostructures, Leioa, Spain. Thank you all for the beautiful and continuous endeavor of driving science and technology a step further together and for sharing this important part of our lives!

Last but not least, we truly thank the team from Wiley for their excellent support: from the first contacts with Rebecca Ralf and Sarah Higginbotham to the last with Shagun Chaudhary, Máire O'Dwyer, Emma Strickland, Rajitha Selvarajan and Lesley Jebaraj, passing through the different colleagues that supported this work; your kindness, patience, continuous support, technical expertise and insights were essential to make this book come true. It has been a real pleasure to work together with you!

Finally, let us hope this first book on printed batteries will promote not only a deeper understanding of this increasingly relevant research and application area but also the interest and motivation to tackle the main challenges, so that we all together contribute to a bright and innovative future in the area of printed batteries!

Carlos Miguel Costa and Senentxu Lanceros-Méndez

1

Printed Batteries: An Overview

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1.1 Introduction

Increasing technological development leads to the question of how to efficiently store energy for devices in the fields of mobile applications and transport that need power supply [1, 2]. Energy storage is thus not only essential but also one of the main challenges that it is necessary to solve in this century [2, 3].

Further, energy storage systems are also increasingly needed, among others, to suitably manage the energy generated by environmentally friendly energy sources, such as photovoltaic, wind and geothermal [4, 5].

Batteries are the most-used energy storage systems for powering portable electronic devices due to the larger amounts of energy stored in comparison to related systems [2, 6]. Among them, the most widely used battery type is lithium-ion batteries, with a market share of 75% [7].

Anode, cathode and separator/electrolyte are the basic components of a battery, the cathode (positive electrode) being responsible for the cell capacity and cycle life. The anode (negative electrode) should show a low potential in order to provide a high cell voltage with the cathode [8–10].

The separator/electrolyte is placed between the electrodes as a medium for the transfer of lithium ions and also to control the number of lithium ions and their mobility [11].

Advances in the area of batteries in relation to printed technologies is expected to have a large impact in the growing area of small portable and wearable electronic devices for applications such as smart cards, RFID tags, remote

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sensors and medical devices, among others. This in fact originated in the development and proliferation of smart and functional materials and microelectromechanical systems (MEMS) needing on-board power supply to provide capacities of 5 to 10 $mAh.cm^{-2}$ with overall dimension of $< 10\,mm^3$ [12–14].

The technological advances of the past years and the need for low-cost and simple processing leads to the potential replacement, in some areas, of conventional processing technologies by printed technologies, as evidenced in applications such as sensors, light-emitting devices, transistors (TFT), photodiodes, flat panel display solar cells and batteries, among others. Printed technology characteristics such as low cost, large area, high volume, light weight, and the processing of multilayered functional structures on rugged and flexible substrates, pave the way for new production paradigms for specific application areas [15–17].

In fact, it is expected that the global market for printed electronics will reach $45 billion in 2017 and is estimated to exceed $300 billion over the next 20 years [15, 16, 18].

This fact is also evidenced by the many articles published in scientific journal about inks and printed electronics, as shown in Figure 1.1.

Printed materials for electronics can be applied on different substrates such as paper, plastics and textiles, giving origin to the term "flexible electronics". Typically, the most frequently used printing techniques for printed electronics are ink-jet and screen-printing [19], but related cost-efficient and high-throughput production techniques such as solution-processing techniques

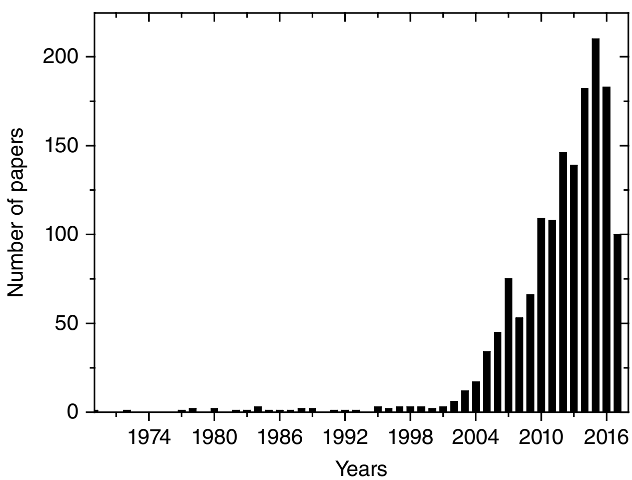

Figure 1.1 Research articles published related to inks and printed electronics. Search performed in Scopus database with the keywords "inks" and "printed electronics" on 19 June 2017.

including spin, spray, dip, blade and slot-die have been used, as well as gravure, flexographic and offset printing technologies [20, 21].

The different printing techniques require the use of specific inks with accurate control of viscosity and surface tension, among other things [22, 23]. Further, for specific printing techniques, the ink properties should be adjusted taking into account the specific pattern to be printed [24].

Printed electronics requires the use of different types of inks such as dielectric, semi-conductive or conductive, which are used to print the different active layers of the devices. Further, inks with piezoelectric [25], piezoresistive [26], and photosensitive [27] properties, among others, have been developed for the fabrication of sensor devices. Typically, inks can be defined as colloidal solutions as the result of a dispersion of organic and/or inorganic particles with specific size into a polymer solution [28]. Moreover, these inks must be cheap, reliable, safe to human health, and processable at temperatures below 50 °C. Further, the inks should preferentially show mechanical robustness, flexibility and recyclability [29].

Independent of the printing process, the ink should be distributed on the substrate with a specific pattern in a reproducible way, which strongly depends on its rheological properties [30].

The rheological properties (flow behavior, flow time and tack) of the ink can be evaluated by using the rotational viscosimeter to measure the viscosity as a function of shear rate, as the material is subjected to multiple shear rates during material processing.

In particular, it is important to prevent the agglomeration or sedimentation of the particles through attractive/repulsive forces, which depends on processing shear rate, as this will strongly affect the final properties of the printed layer [31].

At low shear rate, the viscosity of the inks is higher due to the attraction between particles, which induces their flocculation and immobility. At higher shear rates, the viscosity of the inks decreases through the low flocculation and higher mobility of solvent entrapped between particles [32, 33]. However, the viscosity of printing inks is not only a function of the shear stress but also of time, which plays an important role in the flow process of the ink for each printed element [30].

Further, the physical and chemical stability of the inks is affected by the different fabrication steps (stirring, dispersion, etc.), in which the energy input and mixing time influence both particle stability and degree of dispersion [34].

The combination of printing and battery technologies gives rise to printed batteries; for this at least one of the components should be processed and deposited through printing techniques in order to keep that designation [12, 35].

Figure 1.2 shows the origin of the denomination and the main applications of printed batteries.

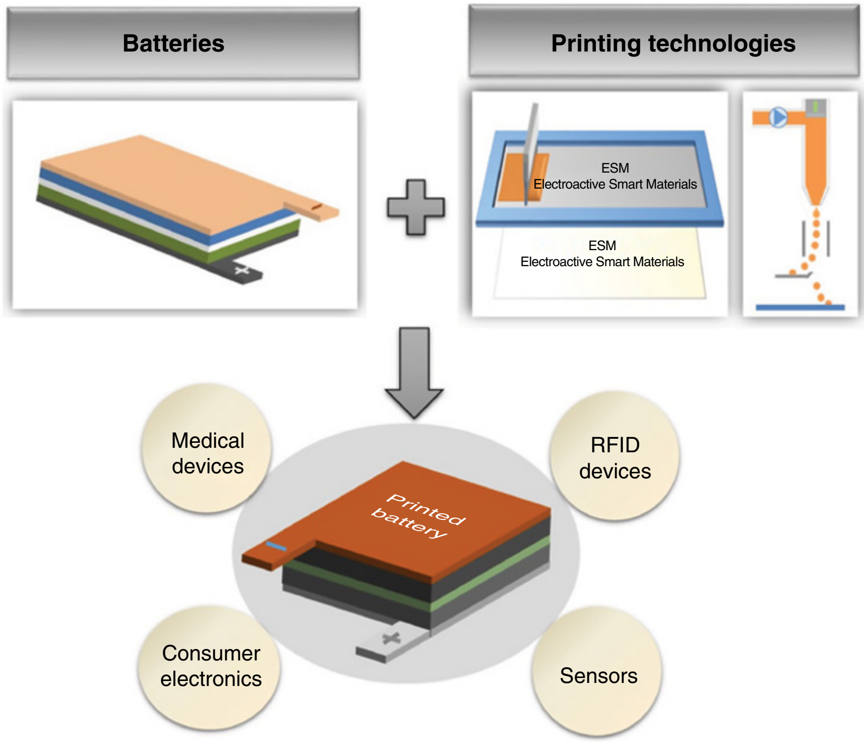

Figure 1.2 An overview of printed batteries and main applications. (*See insert for color representation of the figure.*)

Further, flexible/stretchable batteries [36, 37] and solid-state microbatteries [38] can be included within the printed battery area when one or more components are produced by printing technologies. In addition, there are usually non-printed components such as the current collector, which also serves as support for the printed structure.

Inks for printed batteries are typically composed of a polymer binder, a solvent and suitable fillers, depending on the layer type: electrodes and separator/electrolyte [35]. Suitable fillers are in the form of micro/nanoparticles, nanoplates, nanowires, carbonaceous matter or ionic liquid, among others [29]. The proper transfer of the ink from the printing plate to the substrate is the main function of a printing process [30].

In the field of printed batteries, ink rheology is one of the key issues, due to the high active material loading that may be necessary for proper battery performance. This ink rheology depends mainly on particle size, solid loading concentration and solvent type [39, 40], with adequate ink showing moderate viscosity and weak sedimentation behavior resulting in an homogeneous particle system within a polymer network [31].

The main printed battery component is the electrode (anode and cathode) [22], and different inks have been reported in the literature based on different active materials such as lithium cobalt oxide ($LiCoO_2$) [41] and lithium iron phosphate ($LiFePO_4$) [40] for the cathode, and graphite [42], mesocarbon microbeads (MCMBs) [43] and tin oxide (SnO_2) for the anode [44]. The active material content of the electrode affects its thickness, which in turn influences battery capacity: increasing electrode thickness leads to mass transport limitations of lithium ions in the electrolyte phase leading to a reduction in the capacity of the cell [45, 46]. Also the porosity of the electrodes has a strong impact on battery performance as it influences the effective electronic and ionic conductivity values [47].

On the other hand, the separator/electrolyte has not been printed very often due to the necessary low ionic conductivity, which leads to the use of composite gel electrolytes to achieve ionic conductivity values closer to those of conventional electrolytes [35]. The separator/electrolyte component of printed batteries is mainly based on composite gel electrolytes where the separator layer is soaked in an organic liquid electrolyte (salt dissolved into an organic solvent or ionic liquid to produce an ion-conducting solution in an inert porous polymeric membrane) in which it is important to control the swelling process [35, 48, 49].

Thus, one of the largest challenges is the development of inks for printing solid-state separator/electrolytes with a minimum ionic conductivity of 10^{-4} S/cm and mechanical and thermal stabilities [50].

The efforts and challenges involved in developing and optimizing specific inks for the different battery components that meet the requirements of efficiency, stability and processability for different printed techniques (Figure 1.3) are the main focus of the present fundamental and applied research efforts in this field.

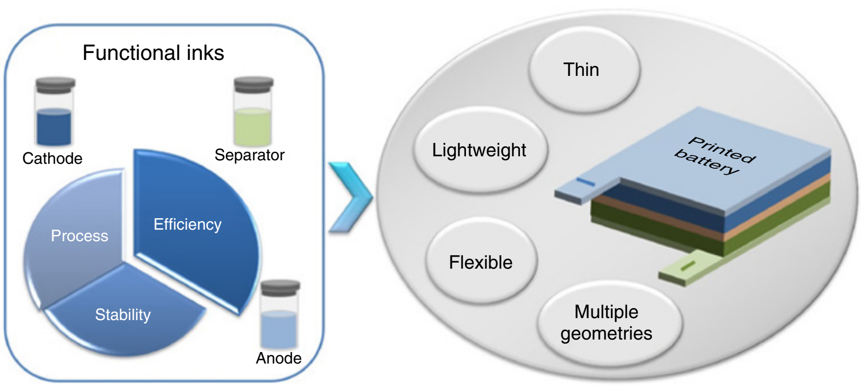

Figure 1.3 An overview of the functional inks and relevant requirements in the area of printed battery research. (*See insert for color representation of the figure.*)

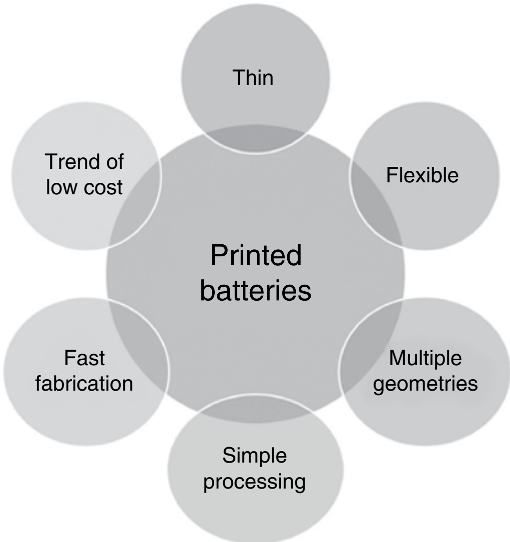

Figure 1.4 Main features and attributes of printed batteries.

The key features and attributes of printed batteries are that they are: customizable, thin, high power, low cost, mechanically flexible, lightweight and rechargeable and that they allow large printed areas. These features will allow the fabrication of functional systems with batteries already integrated in devices [51].

These features and attributes are shown in Figure 1.4 and are the main advantages in comparison to conventional batteries.

The production costs and processing steps for printed batteries can be reduced through the use of roll-to-roll production methods, as they enable the fabrication and assembly of the different layers of the batteries at high speed in a continuous process [52].

Some of the main differentiating factors of printed batteries in comparison to conventional batteries are their simple integration into devices, the possibility of production for large areas and the possibility of thickness reduction. Further, eco-friendly processes and materials are also possible [53].

Currently, research efforts are focused on improving performance (specifically energy and power) and on developing new fabrication processes, inks, designs and characteristics for applications such as smart cards, radio-frequency-identification (RFID) security and information devices, thin-film medical products, and new applications including e-labels, e-packaging, e-posters and medical disposables [54].

A common factor for many of the aforementioned products is the requirement of on-board battery power supply at the microwatt-level with specific designs, which can be achieved with printed batteries.

The performance parameters (i.e., power and energy value, lifetime and discharge rate) of printed batteries are established according to the application, the delivered capacity being between 0.7 and 90 mAh for commercial printed batteries [35, 55, 56].

Thus, printed batteries are being applied in an increasing number of applications and the advances in printable ink formulations and printing technologies, such as 3D-printing, will allow the fabrication of fully printed batteries with high areal energy density to widen the range of possible application areas.

1.2 Types of Printed Batteries

Electrochemical power sources, defined as batteries, were invented by Alessandro Volta, professor at the University of Pavia, Italy, in 1800, and are nowadays an essential component of the electronic devices market as well as of hybrid electric vehicles (HEVs) and electric vehicles (EVs) [57]. The "voltaic pile" consists of an alternating sequence of two different metal discs (zinc, Zn, and silver, Ag) separated by a cloth soaked in a sodium chloride solution [57]. Over the years, different battery types, including Zn, nickel-cadmium and nickel-metal hydride, were developed, with lithium-ion batteries now the most advantageous type.

The first Li-ion batteries were commercialized by Sony in 1991 based on the pioneering work of Yazami regarding the use of lithium-graphite as a negative electrode [58].

Some of the main advantages of lithium-ion batteries include being light, cheap, environmentally friendlier and safer as well as showing higher energy density, less self-discharge, no memory effect, prolonged service-life and higher number of charge/discharge cycles [9, 57].

Batteries are usually defined as primary and secondary, the latter being rechargeable batteries [59, 60]. Independently of the battery type, their main constituents are the two electrodes, anode and cathode, and the separator/electrolyte, as shown in Figure 1.5.

The two main processes of rechargeable batteries are charging and discharging, as illustrated in Figure 1.5. During the charging process, the movement of ions is from the cathode to the anode electrode and during the discharge, the movement is in the opposite direction, i.e., from the anode to the cathode [60].

In Figure 1.5, graphite represents the anode material and lithium-manganese oxide, $LiMn_2O_4$, represents the cathode material.

With respect to printed batteries, the most frequently used ones are based on lithium, Li, and zinc, Zn.

Lithium printed batteries are lithium-ion with different electrodes (graphite or Li_xC_n for anode and $LiCoO_2$, $LiMnO_2$, or $LiFePO_4$ for cathode, lithium-manganese dioxide, $Li\text{-}MnO_2$ and post Li, i.e., lithium-air, sulphur-cathode, etc.)[53].

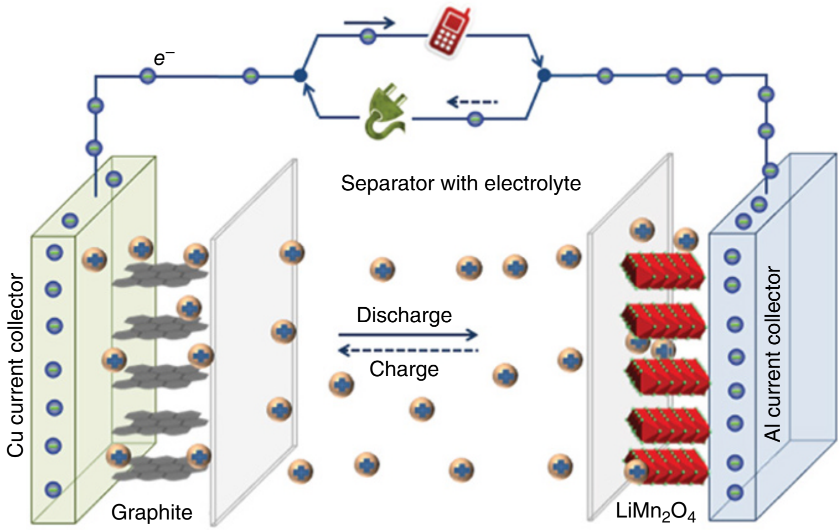

Figure 1.5 Schematic illustration of the main constituents and representation of the charge and discharge modes of a battery. (*See insert for color representation of the figure.*)

In relation to zinc batteries, the most frequently used are zinc-manganese dioxide, Zn-MnO_2 (Zn for anode and MnO_2 for cathode), zinc-air and zinc-silver oxide, Zn-Ag_2O [53]. Further, there are other electrochemical systems, such as nickel/metal hydride, which have been also applied in printed batteries [61].

As an example, for anodes based on graphite and cathodes based on $LiCoO_2$, the rechargeable electrochemical reaction of a lithium-ion printed battery system is:

$$LiCoO_2 + C_6 \underset{disch\arg e}{\overset{ch\arg e}{\rightleftarrows}} Li_{1-x}CoO_2 + Li_xC_6 \tag{1}$$

Typically, zinc battery types are non-rechargeable systems except for nickel-metal hydride. For the Zn-MnO_2 system, which is the most often used in printed batteries, the electrochemical reaction is:

$$Zn + 2MnO_2 + H_2O \rightarrow ZnO + 2MnO(OH) \tag{2}$$

For many small device applications in which no high voltage is required, Zn-MnO_2 batteries can be the most appropriate due to their high energy content, lower internal resistance, large shelf-life and the low cost of Zn and MnO_2 in comparison with lithium battery materials [62].

In relation to the other printed battery types, those that stand out in the literature after Zn-Mn_2O and lithium batteries are Zn-Ag_2O batteries with 1.3 to 5.4 mAh.cm^{-2} at 1.5 V [63, 64].

1.3 Design of Printed Batteries

For conventional batteries, there are basically four main designs, which are coin cell, prismatic, spiral wound and cylindrical [65]. All of them have in common the fact of being rigid and bulky, and not adequate for flexible electronics devices.

One advantage of the use of printing technologies in the fabrication of batteries is that it is possible to develop one or more layers with a specific pattern, i.e., design [66]. This is particularly relevant as together with the characteristics of the materials used for the fabrication of a battery, the geometry/architecture of the battery strongly affects its performance [67]. For printed batteries, the main types are the stack or sandwich architecture (Figure 1.6) and the coplanar or parallel architecture (Figure 1.7).

Figure 1.6 shows the stack or sandwich architecture, which consists of a current collector for the anode, anode, separator with electrolyte, cathode, and current collector for the cathode, all deposited in a flexible substrate with an overall thickness of 0.5 mm for the printed battery.

This architecture is identical to that of conventional batteries; it leads to low internal resistance due to the small distance that the lithium ions travel by moving between the anode and cathode, which also allows shorter charging times.

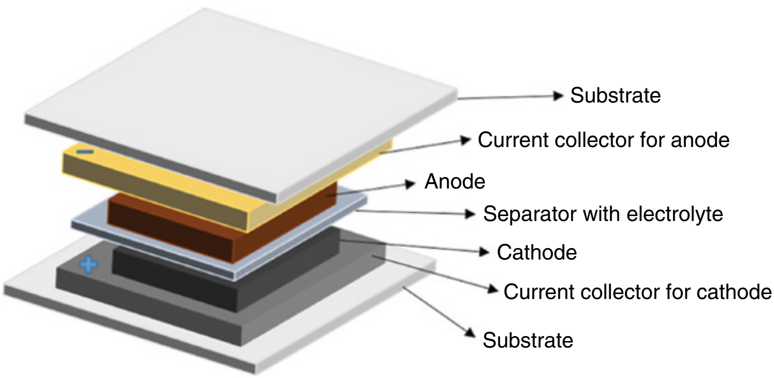

Figure 1.6 Schematic representation of a printed battery in the stack or sandwich cell architecture.

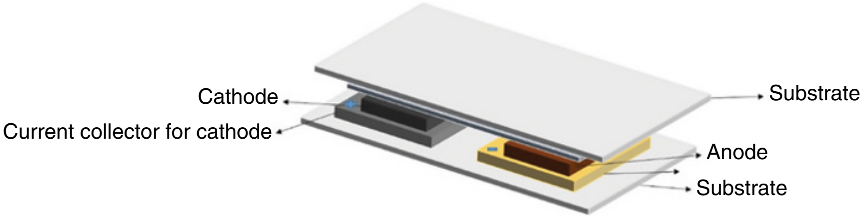

Figure 1.7 Schematic representation of a printed battery in the coplanar or parallel cell architecture.

Figure 1.7 shows the coplanar or parallel architecture for printed batteries, which consists of the anode and the cathode in a side-by-side position. This architecture is the most frequently used for stretchable batteries [68, 69]. In this geometry it may be not necessary to put the separator within the cell [53].

In this architecture, the risk of shorting during battery mechanical stretching is minimal.

It should be noted that, independently of the battery architecture, the sealing process is an essential step in printed batteries. This process consists of a sealing layer based on a polymer glue, which can be processed by the application of heat or pressure, with the main objective of protecting the battery against atmospheric gas molecules such as H_2O and O_2 [53, 70].

Energy storage within the sandwich architecture (Figure 1.6) has been increased by the development of interdigitated architectures, such as the one represented in Figure 1.8 [71]. The interdigitated architecture is based on electrode digits separated by an electrolyte, allowing increased surface area for the electrodes. In this architecture, the Li^+ transport paths are shorter, reducing the electrical resistances across the battery [72]. Further, the ohmic drop of the interdigitated architecture is lower due to the smaller electrolyte/separator layer, leading to increased power.

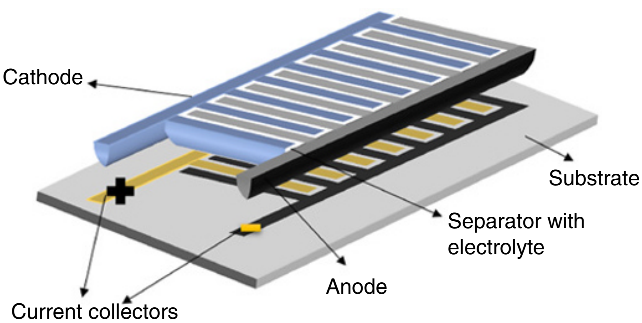

Figure 1.8 Schematic representation of the interdigitated battery architecture.

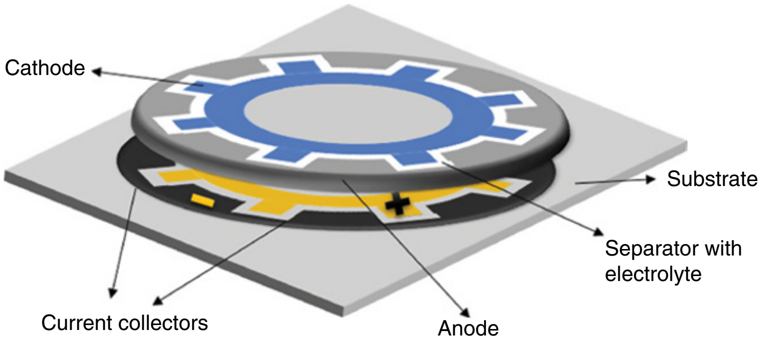

Figure 1.9 Schematic representation of the gear architecture.

Several works have been reported on interdigitated architectures fabricated by printing technologies, such as screen-printing, ink-jet printing and 3D-printing [73, 74].

Printed batteries based on interdigitated architectures have been fabricated based on lithium inks for anode, $Li_4Ti_5O_{12}$ (LTO), and cathode, $LiFePO_4$ (LFP), with high energy density, 9.7 J cm^{-2}, at a power density of 2.7 mWcm^{-2} [74], values compatible with their use in microelectronics and biomedical devices.

The combination of printing technologies and batteries results in novel architectures, customizable for specific applications. In this sense, Figure 1.9 shows a "gear architecture", recently proposed, resulting from the application of the interdigitated architecture to circular batteries. This geometry is suitable for smartwatches, mobile phones and medical devices, among other applications [75].

Thus, printed batteries allow the development of novel architectures with optimized performance and better integration into specific devices [75].

1.4 Main Advantages and Disadvantages of Printed Batteries

1.4.1 Advantages

Printed batteries cannot compete with conventional batteries in applications where there are no size and shape limitations. On the other hand, they can fill the gap for small portable devices in which size, weight and improved integration in the device are some of the main requirements [35]. Other relevant areas for printed batteries are devices in which flexibility and stretch ability are required.

Other main motivations for the implementation of printed batteries is to reduce production costs and/or to achieve specific design features that printing technologies can provide [35].

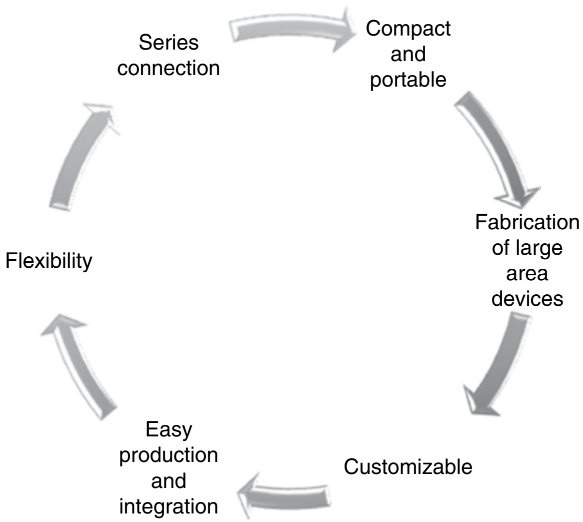

Figure 1.10 Main advantages of printed batteries.

The main advantages of printed batteries are summarized in Figure 1.10.

The main advantages of printed batteries are: easy production and integration; the fact that they are compact and portable, flexible and customizable; and the fact that they can be printed in series connection and allow fabrication of large-area devices (Figure 1.10). Printed batteries can be thinner than a millimeter, lighter than a gram, mechanically flexible and stretchable, allow specific designs and involve cost-effective production on a large scale [35].

One of the relevant advantages of printed batteries is the series connection of cells through printing technologies. In fact, the fabrication of printed batteries up to 15 cells has already been demonstrated [53].

1.4.2 Disadvantages

At the present moment, the higher cost of printed batteries in comparison to conventional batteries is one of the main drawbacks hindering the growth of their market share. Other problems are the need to develop new functional materials (inks) and to optimize processing.

It is expected that as production volumes increase and technology improves, the cost of printed batteries will decrease and they will compete with conventional batteries in price [76].

Figure 1.11 summarizes the current main disadvantages of printed batteries.