Preface

For the entire duration of human history, man has sought sources of energy that can be used to improve their work and lifestyle. Indeed, since prehistory initial rudimentary researches were focused on the use of human/animal forces or the combustion of wood or, in general, of biomass for the production of work. Going forward with time, many other sources of energy were exploited, such as wind and water to move the blades of the mills.

Nowadays, the presence of numerous sources makes the energy available in large quantities and at low prices. This enabled significant infrastructure development and a sharp acceleration of the industrialization process. On the other hand, the evolution of society is leading to an increasing need of energy, thus it is necessary to evaluate all the problems related to this enormous demand.

In this context, it is evident that the necessity to find abundant, clean and cheap energy sources is one of the priorities of the twenty-first century. Currently, the energy demand is fulfilled almost only thanks to fossil fuels and, to a lesser degree, by nuclear power. Various technologies are under development to use renewable sources readily available in nature, such as wind and solar energies, waste biomass combustion products, geothermal energy or the one coming from the motion of sea currents. Nevertheless, the sources of sustainable energy are intermittent and/or restricted in specific areas; as a result, they require the use of suitable technologies for energy storage.

After these considerations, it is clearly evident that reliable methods for storing energy are fundamental and secondary lithium-ion batteries have all the characteristics to be one of the most attractive solutions. The present battery technology based on lithium outperforms many other conventional systems, such as the lead-acid, nickel-cadmium and nickel-metal hydride batteries, because of its high energy and power density. The Li-ion battery is a compact, lightweight, rechargeable power source stable to over 1000 charge/discharge cycles. It can be fabricated in size ranging from few microns to a large-scale battery capable of providing power for computer memory chips, communication equipment, colour motion pictures and, potentially, for the huge market of electric vehicles (EV) and
hybrid-electric vehicles (HEV), where low-cost, low environmental impact, as well as high-specific performance batteries are needed.

The Li-based battery chemistry is well established, but still relatively young. Thus, several improvements can be made: optimization of existing chemistries by the introduction of environmentally friendly materials and the simplification of the device production process are intriguing challenges. Moreover, the recent developments in the next-generation electronic devices promoted the modification of the current systems towards lighter, more flexible and/or micro-sized ones. The enhancement of the mechanical properties through the introduction of flexible electrodes would enable lithium-based batteries to be embedded into a wide variety of innovative products such as smart cards, wearable displays and implantable medical devices. Moreover, the optimization of the electrolyte substituting the liquid electrolyte with a solid membrane would offer adaptability to various designs and stressful mechanical handling, as well as greatly improve the overall cell safety.

Starting from the above-discussed considerations, the research work I made during my Ph.D. was focused on the use of natural cellulose fibres for the production of innovative, low-cost and easily recyclable Li-ion cells by means of fast and reliable paper-making procedures exploiting water as solvent and was performed under the supervision of Prof. Penazzi, Prof. Gerbaldi (Politecnico di Torino) and Dr. Beneventi (LGP2—Pagorà Grenoble); moreover, some specific methods are proposed for the optimization of the safety features of the paper-based cells as well as for the improvement of the electronic conductivity of the electrodes to achieve high rate capabilities. In order to successfully reach the proposed challenging goals, the research work was carried out in the collaboration with the laboratories of LGP2—Pagorà in Grenoble (France) where I spent a period of 18 months.

The first three chapters of the monograph represent the introductory section that should provide the general information to understand the manuscript. In particular, Chap. 1 deals with the basic concepts for cells and batteries and with a brief description of the general characteristics of the mature portable power source technologies.

Chapter 2 discusses the present status and future trends in Li-based batteries research as well as their main characteristics, working principles and components.

Chapter 3 describes the main properties, uses and production methods for both cellulose and cellulose derivatives. A brief review about the use of cellulose in electrochemical devices is also presented.

In Chaps. 4 to 8 the experimental results obtained during my research work are thoroughly discussed.

Chapter 4 describes the materials, instrumentations, methods and procedures used during the work.

Chapter 5 describes a procedure tuned by Jabbour et al. for the production of Li-ion battery electrodes exploiting filtration. This procedure allows to substitute the commonly used synthetic binders with truly natural cellulose and the harmful organic solvents with water. My work here consisted in the optimization of the
starting slurry formulations and in electrochemical testing of the resulting electrodes.

In Chap. 6 an innovative, fast and reliable process that couple electrochemical knowledge with paper-making technology is presented. In such an innovative procedure, a spray coater for the deposition of the active materials was added on a pilot line for the production of common paper sheets with the aim of obtaining lithium battery electrodes on a preindustrial scale.

Chapter 7 is focused on the development of the third fundamental battery components, which is the separator. In the first section several studies were carried out on the development and optimization of paper handsheets to be used as separators. In the second section a methacrylic-based composite polymer membrane produced with the method proposed by Chiappone et al. and using microfibrillated cellulose as reinforcing agent was coupled with the spray-coated paper-based electrodes resulting in an all-paper-based quasi-solid Li-ion cell showing interesting electrochemical performances and stable cycling characteristics.

Finally, in Chap. 8 a carbonization procedure is demonstrated, which specifically leads to the enhancement of the electronic conductivity of the spray-coated paper-based electrodes; this, in turn, results in an increased power capability of paper-based Li-ion cells making them suitable for more power demanding systems. Noteworthy, due to the very high electronic conductivity, the carbonized paper sheet may successfully replace the metal current collector foil, thus giving the possibility to produce light and mechanically abusable Li-ion cells to be implemented in the next generation of flexible electronic systems.
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