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Cheap and easily processable electrode/electrolytes for next-generation sodium-ion batteries

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Abstract. Electrochemical energy storage is of increasing importance to allow large-scale integration of intermittent renewable sources. State-of-the-art lithium-ion batteries suffer from relevant problems, including strategic materials supply (e.g., Li, Co), high environmental impact, high cost, low safety. Cheaper and safer batteries based on sodium-ion chemistry can answer to these issues. In this work, we demonstrate how we aim at developing new materials for next-generation, highly performing and sustainable, all-solid-state, secondary Na-ion batteries. We will explore the entire value chain, starting from the development of carbonaceous electrodes obtained by pyrolysis of biosourced waste to the development of advanced, polymeric electrolytes, also including computationally-assisted investigation of electrode/electrolyte materials and their “green” fabrication and assembly/testing in lab-scale cells.

Key words. energy storage, sodium-ion battery, carbonaceous anode, solid polymer electrolyte, electrochemical characterisation.

1. Introduction

Energy storage is a key challenge of the 21th century. In fact, renewable energy production is expected to grow largely in the coming years, and efficient massive storage is required to improve large-scale grid integration of intermittent electricity sources (e.g. solar, tides, wind) [1]. At present, the state-of-the-art in the field is represented by Li-ion batteries. However, near future global battery market might be so large that problems regarding materials supply will rise not only for lithium (*viz.* Li₂CO₃), but also for electrode raw materials (e.g. Co-based). Cheaper batteries based on Na-ion chemistry can refresh the renewable energy sector and supply more balancing to the grid, providing proper back up to intermittent renewable sources. Sodium exhibits suitable electrochemical properties, close to those of lithium, and it is very abundant. These features make Na-based batteries proficient candidates to replace LiBs, particularly when large-scale energy storage is envisaged [2]. This chemistry

might be strategic for electrified road transportation as well, which has stringent requirements in terms of safety and durability. In general, development of new Na-ion based technologies requires: i) the discovery/investigation of new materials/components being widely distributed in large amount and without strategic contingencies, and ii) the search for high energy/power density, and safe battery configurations. In this context, in this work we will deliver a fully integrated innovation chain based on the material-to-end-use approach. In particular, the target in our research group is to develop new materials for next-generation, highly performing and sustainable, all-solid-state, secondary Na-ion batteries, by exploring the entire value chain that includes “green” fabrication of nanostructured electrode (e.g., iron and/or vanadium phosphates eventually supported on electrospun carbon fibers at the cathode and biosourced derived hard carbons at the anode) and solid electrolyte materials (crosslinked polyethyleneoxide with specific plasticisers and additives), and their assembly and electrochemical testing in lab-scale test cells with the best materials, validated upon prolonged cycling at ambient temperature. Concerning sustainability, recyclable and naturally abundant materials are processed to give energy storage components with minimum environmental impact.

2. Overview of the research work

Below, new materials for next-generation, highly performing and sustainable, solid-state Na-ion batteries under development in our labs are briefly detailed.

A. Nanostructured electrode materials

Different kind of electrode materials for Na-ion batteries are under development in our laboratories, which include sodium iron phosphate specifically mixed with reduced graphene oxide or vanadium phosphates eventually supported on electrospun carbon fibers at the cathode and

biosourced-derived hard carbons with graphenic domains at the anode. Phosphate materials are considered at the cathode because of their high structural stability and the possibility of tuning the working potential by modulating the metal content into the phosphate framework. Cathode materials are developed by different synthetic routes and sampling different metal compositions. To overcome the intrinsic limitation of poor electronic properties, nanostructured powders are mixed with high surface carbon composites, such as in particular reduced graphene oxide, eventually exploiting a spray-drying procedure, or supported (by simple filtration) on self-supported nanosized carbon fibers (CNF) obtained by carbonisation of electrospun polyacrylonitrile nonwovens (Figure 1).

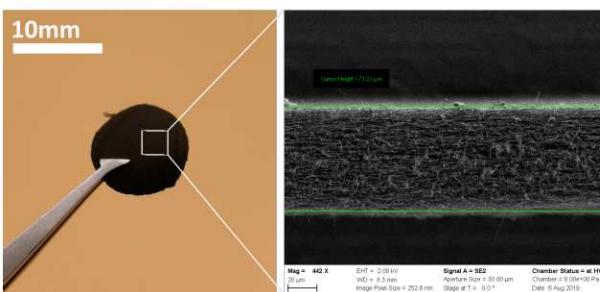


Fig.1. SEM images of the CNF support for cathode material.

At the anode side, we rely on the Na intercalation into carbonaceous matrixes with turbostratic structure, bearing alternated amorphous and crystalline domains of graphene. By optimizing precursors and syntheses, we can tune graphenic domains features critical for the electrochemical performance. Enhancements beyond the state-of-the-art concern long-term stability (target >1500 cycles) and high capacity (>450 mAh/g @100% DoD). Innovative generations of carbon-based composite electrodes enabling flexible, low cost and current-collector free technology are developed based on self-standing carbon paper [3]. Here we cover the whole value chain from the supply of the materials: abundant and cheap waste deposits both from daily consumables and agricultural residues are expected to assure industrial level production with low environmental impact.

B. Solid-state polymeric electrolytes

Polymer electrolytes are prepared through different techniques, exploiting both simple casting and UV-curing and using various additives to improve specific characteristics (e.g., RTILs, cellulose derivatives, glymes) [4,5]. In particular, UV-curing allows obtaining self-standing polymer electrolytes with desirable properties of flexibility, shape retention upon thermal stress, improved interfacial contact with the electrodes and ionic conductivity suitable for practical application. They exhibit excellent ionic conductivity and wide electrochemical stability window, which ensures safe operation at ambient conditions. Electrochemical performances in lab-scale devices are presented, evaluated by means of cycling voltammetry and galvanostatic charge/discharge cycling exploiting different nanostructured electrode materials (e.g., hard carbons, TiO₂, high voltage phosphates prepared by water-based procedures with green cellulosic binders).

The highly ionic conducting (>1 mS cm⁻¹ at 25 °C) and self-standing (Figure 2) polymer-based electrolytes are used in lab-scale Na cells.

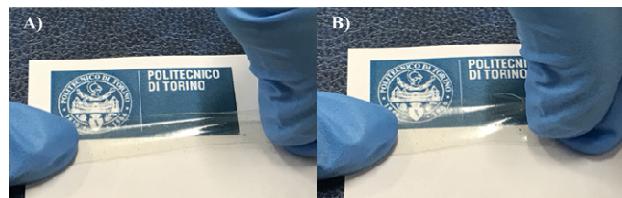


Fig.2. Digital photographs of a crosslinked polymer electrolyte. A) under stretch and B) in relaxation mode.

The obtained results in terms of ambient temperature cycling behaviour (stable specific capacity exceeding 250 mAh g⁻¹ at 0.1 mA cm⁻² and overall remarkable stability, for a quasi-solid state Na polymer cell, upon very long term cycling exceeding 1000 reversible cycles at 0.5 mA cm⁻² corresponding to > 5000 h of continuous operation) demonstrate the promising prospects of the novel crosslinked polymer electrolyte under development to be implemented in the next-generation of Na-based batteries conceived for large-scale energy storage systems, such as those connected to photovoltaic and wind factories.

3. Conclusion

Work on Na-ion polymer batteries for moderate temperature application is at an early stage, only lab-scale cells were demonstrated so far. Nevertheless, with the appropriate choice and optimization of electrode/electrolyte materials (and successful combination thereof), the intriguing characteristics of the newly developed systems here presented postulate the possibility of their effective implementation in safe, durable and high energy density secondary Na-based Na-based energy storage devices conceived for green-grid storage and operating at ambient temperatures in solid-state configuration.

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