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Reconfigurable sticker label electronics manufactured from nanofibrillated cellulose-based self-adhesive organic electronic materials

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ABSTRACT

Low voltage operated electrochemical devices can be produced from electrically conducting polymers and polyelectrolytes. Here, we report how such polymers and polyelectrolytes can be cast together with nanofibrillated cellulose (NFC) derived from wood. The resulting films, which carry ionic or electronic functionalities, are all-organic, disposable, light-weight, flexible, self-adhesive, elastic and self-supporting. The mechanical and self-adhesive properties of the films enable simple and flexible electronic systems by assembling the films into various kinds of components using a “cut and stick” method. Additionally, the self-adhesive surfaces provide a new concept that not only allows for simplified system integration of printed electronic components, but also allows for a unique possibility to detach and reconfigure one or several subcomponents by a “peel and stick” method to create yet another device configuration. This is demonstrated by a stack of two films that first served as the electrolyte layer and the pixel electrode of an electrochromic display, which then was detached from each other and transferred to another configuration, thus becoming the electrolyte and gate electrode of an electrochemical transistor. Further, smart pixels, consisting of the combination of one electrochromic pixel and one electrochemical transistor, have successfully been manufactured with the NFC-hybridized materials. The concept of system reconfiguration was further explored by that a pixel electrode charged to its colored state could be detached and then integrated on top of a transistor channel. This resulted in spontaneous discharging and associated current modulation of the transistor channel without applying any additional gate voltage. Our peel and stick approach promises for novel reconfigurable electronic devices, e.g. in sensor, label and security applications.

1. Introduction

Since its discovery about 2000 years ago, papers are one of the most commonly manufactured and utilized sheet materials in our world; it represents the planar carrier that we utilize to record, transfer and share printed information. In fact, it is the largest surface ever manufactured by mankind. During the digital revolution, paper has certainly been challenged. However, nowadays, rather thanks to the digital revolution the interest for paper as the carrier for information is regained. One of the prime reasons for this revival is the birth of the technology and science field

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known as printed electronics. From an application point of view paper now turns into the carrier for flexible electronics, systems that can be manufactured using the same printing tools that were originally developed for graphics and texts. Printed electronics defined on paper labels, package board and on fine paper, is expected to extend the function of paper and to make paper products connected to the digital world.

Paper is based on wood fibers comprising the most abundant organic compound derived from biomass, namely cellulose. One such renewable cellulose material, nanofibrillated cellulose (NFC), has attracted a very large interest during the last decade. This material emanates from the cellulose fibrils inside the wood fiber wall and the fibrils are liberated in different ways using both chemical treatment and high-pressure homogenization to separate the fibrils from each other and to stabilize them in aqueous dispersion [1,2]. The attraction of NFC stems from their interesting intrinsic properties such as its high specific surface area, flexibility, high aspect ratio of the fibrils, good mechanical properties, and its film forming capacity. NFC dispersions converted into "nanopaper" films can be transparent, reach mechanical properties similar to that of cast iron [3], or show very good oxygen barrier properties. NFC has also been used as a filler to provide reinforcement, flexibility and transparency in nanocomposites and displays [4,5].

The field of printed electronics is presently attracting a lot of interest since it promises for distributed intelligence and monitoring; features that are expected to combat some of the challenges in our society related to health care, monitoring, logistics and safety. A tremendous variety of components have been developed and manufactured on flexible substrates using different kinds of coating, printing and lamination tools [6–8]. Examples include batteries, capacitors, solar cells, displays, diodes, transistors and logic circuits. To achieve true printed electronics applications, typically several different kinds of components need to be integrated into a monolithic flexible system. However, integration of printed electronic subcomponents has proven to be a major challenge with respect to the merger and compatibility of different printing technologies, materials and post-processing steps. In fact, in many cases specific manufacturing steps are mutually exclusive since some steps tend to destroy or deteriorate features and components manufactured earlier in the integration scheme. This then implies a decreased production yield that can only be solved by typically increasing the cost for production and integration. In addition to this, the complexity of such production process also brings along the requirement of huge investments of non-standard manufacturing equipment. The overall mindset when developing the printed electronics platform must therefore aim at minimizing the number of materials and process steps, as well as trying to make the processes of different subcomponents compatible with each other by utilizing the same kind of materials for different functionalities [9].

One of the major advantages of using electrochemical devices, based on for instance an electrochemically active conjugated polymer and an electrolyte, is that the very same material can be used for a versatility of device functions. The very same electrolyte component can be used both as the gate insulator in electrochemical and electrolyte-gated field effect transistors, and also as the electrolyte in electrochromic (EC) display devices, batteries and capacitors. PEDOT:PSS, poly(3,4-ethylenedioxythiophene) doped with poly(styrene sulfonate acid), can serve as the conductor, display electrode and also as the transistor channel in printed integrated circuits, such as displays, indicators and sensor systems. The PEDOT:PSS material is electrically conducting and optically transparent in its pristine state and it exhibits electrochromic switch characteristics and control of the conductivity upon electrochemical switching. PEDOT is a π-conjugated electronic polymer and the PSS phase serves as its counter ion [10]. In the neutral state PEDOT appears deep blue [11] and exhibit semiconducting properties. PEDOT can reversibly be switched in between its oxidized and neutral state. The electrochromic effect can be utilized in transmissive displays by using transparent electrolytes, or in a reflective mode of display operation in which the counter electrode is hidden under an opaque electrolyte [12]. The latter version is typically used when PEDOT:PSS serves as both the counter and the pixel electrodes since such configuration maximizes the color switch contrast of the resulting display. However, there are different strategies in order to enhance the color switch contrast also for applications where transmission mode is desirable, for example by using a bottom display electrode and a top display electrode that together express a complementary EC switching characteristics with respect to each other. Polyaniline (PANI) is such an EC material that electrochemically switch color in a complementary fashion with respect to PEDOT, i.e. it exhibits a faint yellow, close to transparent, color in its reduced leucoemeraldine state, while PANI becomes dark blue, almost violet, in its oxidized pernigraniline state. Hence, an electrochromic display comprising a transparent electrolyte layer sandwiched by one PEDOT:PSS electrode and one PANI electrode can switch between a close to transparent pixel state to a dark black-blue colored state, where the latter color state is obtained by applying the positive and negative voltage to the PANI and PEDOT:PSS electrode, respectively [13].

Besides reducing the number of different materials in printed electronics to achieve a resulting robust and rational platform for flexible electronics, one should also consider radically new integration concepts. In packaging and graphic art industry labels are commonly adhered to paper surfaces and products to generate a final integrated system or to extend the functionality of a specific product. Often, these add-on stickers include a coating that provides pressure-sensitive adhesion of the label to the surface of the carrier. In some cases those stickers can later be removed and then transferred to a different new carrier or item to serve yet another application.

Previous attempts on using "peel and stick" or "cut and stick" techniques have resulted in flexible thin film solar cells [14] and free-standing dielectric layers for use in transistors [15], respectively. Additionally, a novel method for integration of memory devices onto flexible substrates has been reported [16], as well as that nanocellulose-based composite materials has been examined for utilization in