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In-situ synthesized polypyrrole-cellulose conductive networks for potential-tunable foldable power paper

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ABSTRACT

Flexible electronics with features of simple, low-cost, lightweight, recyclable, easily operable, and disposable are particularly desirable. Herein, a flexible fully-integrated self-powered system on paper was demonstrated by incorporating addressable paper circuit switch into potential-tunable foldable power paper. In which the electrode material as well as conducting interconnections, p-toluenesulfonic acid (p-TSA) doped polypyrrole embedded in-between the cellulose fibers (p-TSA-PPy/cellulose) were obtained by combining in-situ chemical oxidative polymerization with paper microfluidic technique. In such unique integrated configuration, the utilization of p-TSA-PPy/cellulose network not only dramatically enhanced the horizontal electrical conductivity, but also conquered the issue of vertical electroconductivity, allowing for the top-channel-bottom conductive interconnections. As a proof-of-concept, an all-paper microfluidic analytical platform that integrated paper-based electrochemiluminescence strategy and proposed power paper was constructed with comparable performance. This work greatly improves the understandings of all-paper-based self-powered systems, and presents the significant potential applications of paper-based flexible electronics.

1. Introduction

Nowadays, there has been significant interest in the development of state-of-the-art electronic devices with multiple-functions to meet the demands of practical applications ranging from portable and wearable functional devices [1–3] to medical monitoring systems [4–6]. The emerging requirements for inexpensive, easily accessible electronics calls for flexible, renewable, environmental friendly, and sustainable power sources [7–10]. Notably, rough and surface-absorptive cellulose paper with unique porous architecture, used ubiquitously in our everyday lives, are exactly met the special demand for substrate material. By virtue of its attractive nature of paper, such as, low-cost, lightweight, power-free fluid transport via capillary action, and biodegradable, as well as the simple assembly strategies, paper-based power sources have been promising candidates for the future disposable and green electronics [11–13]. As a consequence, paper-based functional electronics has endowed a new era of applications in energy systems [14–16], radio-frequency identification [10], electronic displays [17], and microfluidic analysis [18,19].

Due to their intrinsic properties and outstanding performance, there is an extensively growing attention focus on paper or paper-like substrates for batteries and other energy storage devices, including electrochemical batteries [20–22], lithium batteries [23,24], biofuel cells [25,26], supercapacitors [14,27–30], and nanogenerators [31,32]. Nevertheless, most of them cannot be considered either environment-friendly or economically disposable, since the electrode materials or electrolyte utilized is unstable, flammable, precarious, or environmentally hazardous [33]. Although nanogenerators could easily and efficiently convert vibration energy in living environment to electrical energy [31,34], but small electronics could not be continuously driven owning to the limited output power. Furthermore, only single voltage output could be obtained because of its intrinsic features of battery, which limit their application to some extent. For instance, traditional batteries must be equipped with potential transformer to trigger quantum dots based electrochemiluminescence (ECL) reaction, resulting in increase of complexity, which, in turn, makes them high-cost tools [35,36]. Therefore, constructing fully-paper-based flexible electronics with suitable power sources remains a significant challenge. Origami, the ancient art of paper folding, has recently been recognized as an effective technique to build up unique architectures via high degrees of folding along predefined lines, such as deformable energy storage devices [20,37–40], and 3D paper-based analytical devices

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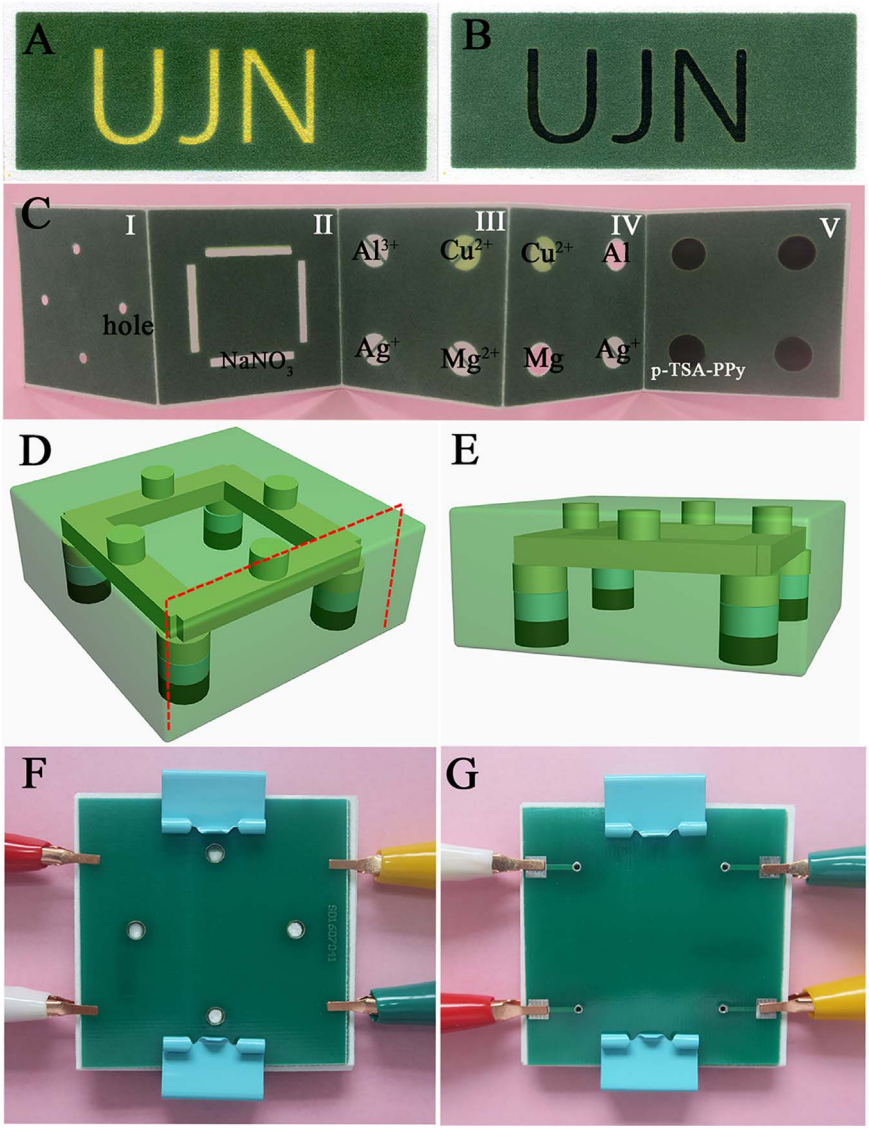

Fig. 1. (A, B) Schematic layout of the in-situ preparation process. Hydrophilic regions of the paper device were filled with $FeCl_3$ (A) before polymerization, then black p-TSA-PPy/cellulose layer (B) were obtained upon the oxidation reaction of Py monomers in the region previously occupied by the oxidant agent. (C) Photograph of a wax printed channels, for forming a potential-tunable foldable power paper, before folding. Layer I with four holes for the injection of water or electrolyte; The microfluidic channels in layer II to IV were filled with $NaNO_3$ solution and four different metal ions, respectively; Two holes in layer IV were cut for two positive electrodes (magnesium and aluminum foils); The black circular regions in layer V were covered by p-TSA-PPy/cellulose. Concept of origami power paper, perspective drawing (D) and side view (E). Photograph of the front (F) and back (G) cover of constructed potential-tunable foldable power paper clamped between the circuit boards.

[41,42]. Inspired by mentioned above, the emerging demand for fully-paper-based flexible electronics could be achieved by incorporating the origami approach.

In the present work, a flexible fully-paper-based self-powered system was demonstrated by incorporating origami skill. In such self-powered system, in-situ synthesized p-toluenesulfonic acid (p-TSA) doped polypyrrole cellulose (p-TSA-PPy/cellulose) conductive networks based on chemical oxidative polymerization with paper microfluidic technique were utilized as electrode material as well as conducting interconnections. Notably, potential-tunable paper-based power sources and addressable paper circuit switch were obtained and incorporated, which greatly facilitate system integration. The generated electrical output by the proposed power paper has been demonstrated to power up commercial light-emitting diodes (LEDs) or trigger ECL sensing platform. The results reveal the feasibility of employing such device as energy components in self-powered system. To the best of our knowledge, there is no report on such p-TSA-PPy/cellulose-based integrated platform up to now. We believe that these findings will pave the way for the rational devise and fabrication of integrated platforms with the distinct features of maximum functionality at minimized size and weight, especially for the development of green electronics.

2. Experimental section

2.1. Design of paper-based electrofluidic device

As one of the vital inventions in human civilization, cellulose papers, used ubiquitously in our daily lives, could play significant roles in the development of flexible and lightweight energy storage [2,43,44]. Whatman chromatography paper grade 2 obtained from GE Healthcare Worldwide (Pudong Shanghai, China) was selected as the substrate material ascribe to its flexibility, light weight, fast water absorption and quick chemical transport rates. Prior to experiment, the size of paper was adjusted with dimensions of 297 mm×210 mm. Then, the patterns of microfluidic channels and functional areas designed by Adobe