Integrated Flexible Solid-state Thin Film Supercapacitors

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In the recent years, there is interest in flexible, thin film solid-state supercapacitors for emerging applications in portable and wearable electronics. Such supercapacitors typically consist of two electrodes sandwiching a solid electrolyte layer. These devices have traditionally been fabricated by making the electrode and electrolyte layers separately, and then holding them together under mechanical pressure[1] or by hot pressing all the layers together[2]. This also promotes adhesion between the electrode material and the solid electrolyte, which can significantly impact the device performance. The mechanical pressure approach limits how supercapacitors are packaged for real-life applications, while hot pressing limits the materials that can be used in the device layers. This work demonstrates a wholly integrated, freestanding thin film supercapacitor, without the need for any mechanical or thermal pressing to hold the device together (Figure 1). We achieve this by a simple and scalable solvent-based fabrication method where active electrode material is directly incorporated on either side of a solid electrolyte layer. We expect our approach to greatly simplify the packaging of thin film supercapacitors for practical applications and promote adhesion between electrode material and solid electrolyte in these devices.

Our approach involves dispersing multiwalled carbon nanotubes (MWCNTs) in a solvent such as isopropyl alcohol (IPA), and casting this dispersion on the surface of a solid electrolyte which is a composite layer of polyvinyl alcohol (PVA) and phosphoric acid (H_3PO_4). The isopropyl alcohol causes swelling in the polymer matrix of the solid electrolyte, causing the MWCNTs to get partially embedded on the electrolyte surface. This makes it possible to achieve an integrated supercapacitor structure, while improving the electrode-electrolyte adhesion. The fully integrated, freestanding, flexible supercapacitors developed in this work show capacitance of more than 2 F/g, with negligible decline in performance over 2000 charge-discharge cycles (Figure 1). Our studies also show a high quality of the electrode-electrolyte interface. Our approach makes it possible to stack such thin film supercapacitors together during the fabrication stage, ultimately allowing for high energy density portable applications.

[1] R. Wang, X. Yan, J. Lang, Z. Zheng, P. Zhang, J. Mater. Chem. A. 2, 12724 (2014).
[2] X. Yang, L. Zhang, F. Zhang, T. Zhang, Y. Huang, Y. Chen, Carbon 72, 381 (2014).

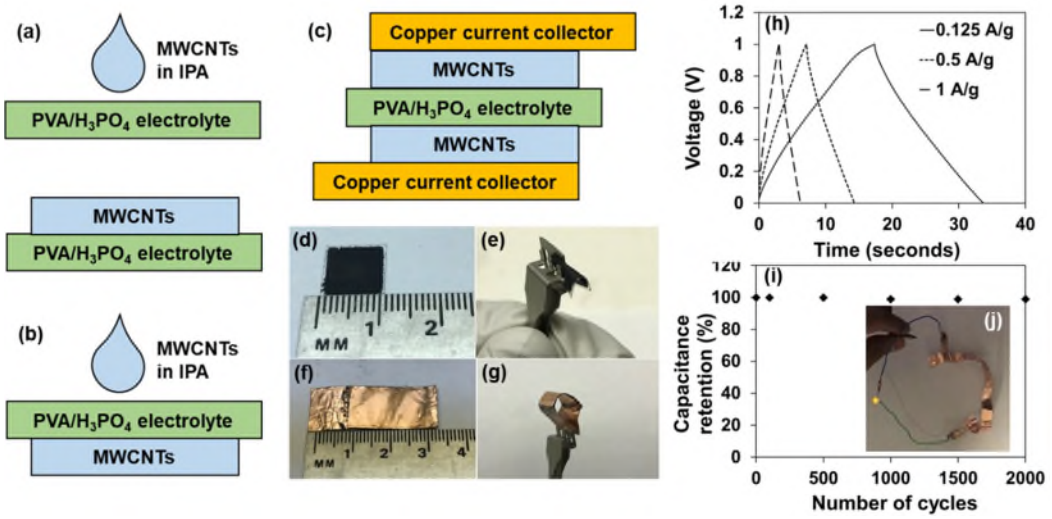

Figure 1: (a) Schematic diagram of solution casting of active electrode material (MWCNTs dispersed in IPA) on one side of a solid electrolyte layer made of polyvinyl alcohol (PVA)/phosphoric acid (H_3PO_4) composite. Upon drying, a solid electrode layer is formed with good adhesion to the electrolyte. *(b)* The process is repeated to form a MWCNT electrode similarly on the other side of the solid electrolyte. *(c)* Schematic of the finished supercapacitor device. *(d), (e)* Images of a fabricated supercapacitor (freestanding and flexed) without current collectors or contacts. *(f), (g)* Images of supercapacitor (freestanding and flexed) after attachment of copper tape as current collector. *(h)* Charge-discharge characteristics of the supercapacitor at different current densities. *(i)* Capacitance retention over 2000 charge-discharge cycles. *(j)* A yellow LED (3.6 mW) powered by the supercapacitor.