

# Improving Interfacial Adhesion Between Active Material and Solid Electrolytes in Thin Film Supercapacitors

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Solid-state thin film flexible supercapacitors are of interest for emerging applications including portable and wearable electronics. Adhesion between the electrolyte (e.g. ionic conductor in a polymer matrix) and the active electrode material (e.g. carbon nanomaterial) is an important factor influencing the performance of the supercapacitor. Typically, discrete electrode and electrolyte layers are fabricated and held together by pressure clamps or mechanical vises to improve electrode-electrolyte interfacial adhesion [1, 2]. However, this approach complicates the packaging of the devices for practical applications. There is a need for simple and scalable alternatives to improve the quality of electrode and electrolyte interface.

We report a facile method for improving adhesion between electrolyte and active electrode material in solid-state supercapacitors. The method involves dispersing active electrode material in a solvent, and depositing the dispersion on the surface of a solid electrolyte. The solvent causes swelling of the polymer matrix of solid electrolyte, which results in the active material being partially embedded on electrolyte surface. This allows for enhanced adhesion between the electrode and electrolyte. The choice of solvent in the electrode material dispersion is influenced by the polymer matrix used in the solid electrolyte. The method reported here has been used to develop a flexible, solid-state thin film supercapacitor. This device was made by solution casting a dispersion of multi-walled carbon nanotubes (MWCNTs) and isopropyl alcohol (IPA) on one side of a solid electrolyte layer (composite of polyvinyl alcohol and phosphoric acid). The IPA swells the crosslinked PVA polymer matrix, and partially embeds the MWCNTs on the electrolyte surface. The process is repeated to fabricate an electrode layer on the other side of the electrolyte layer, resulting in a supercapacitor with an integrated structure. Consequently, there is no need for mechanical clamps to hold the structure together. Devices fabricated in this manner show capacitance of 1.8 F/g, with areal capacitance of 7 mF/cm<sup>2</sup>. The energy and power densities are 0.25 Wh/kg and 1.25 kW/kg respectively. The devices are flexible and do not show appreciable degradation in performance over 2000 charge-discharge cycles and when operating at temperatures as high as 60°C. Electrode-electrolyte interface characterization shows good adhesion between MWCNTs and solid electrolyte.

[1] R. Wang, X. Yan, J. Lang, Z. Zheng, P. Zhang, *J. Mater. Chem. A*, 2, 12724 (2014).

[2] X. Zhang, Z. Lin, B. Chen, S. Sharma, C. Wong, W. Zhang, Y. Deng, *J. Mater. Chem. A*, 1, 5835 (2013).

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## Supplementary Page

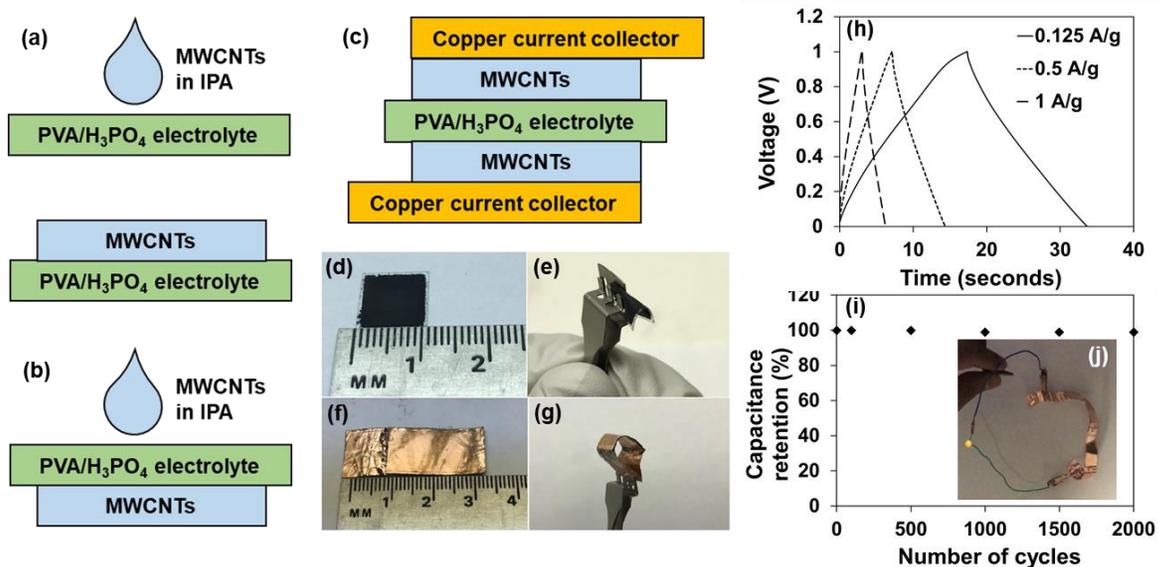


Figure 1: (a) Schematic diagram of solution casting of active electrode material (MWCNTs dispersed in IPA) on one side of a solid electrolyte layer made of polyvinyl alcohol (PVA)/phosphoric acid (H<sub>3</sub>PO<sub>4</sub>) composite. Upon drying, a solid electrode layer is formed with good adhesion 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.