

Flexible Supercapacitor Device based on Free-standing  
Transition Metal Oxides / Reduced Graphene Oxide  
Hybrid Paper Electrode

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mechanical and electrochemical durability of the flexible  
supercapacitor device.

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ABSTRACT

Flexible energy storage devices have attracted significant attention due to their immediate application in the foldable/wearable displays or personal devices. In order to meet the requirement of the flexible gadgets, in addition of having good electrochemical properties, the electrodes of the energy storage devices need to exhibit excellent mechanical robustness and compact design.

Due to their outstanding mechanical and electrical properties as well as notable electrochemical stability, graphene based papers have been introduced as one of the possible candidates for the flexible electrode. However, the capacitance of the free standing graphene based papers is still limited to  $\approx 150 \text{ F g}^{-1}$ . The performance of graphene based paper can be further improved by incorporating pseudocapacitive materials, such as transition metal oxides. Several works on the hybrid paper electrodes based on transition metal oxides/graphene based paper electrodes have been reported. However, stacking and aggregation problem are commonly observed in those hybrid paper electrodes, preventing the optimum electrochemical performance of the hybrid paper.

In this communication, a general strategy has been developed to synthesize transition metal oxide/reduced graphene oxide paper electrodes through in situ and low temperature synthesis method. The versatility of the method has been tested on several types of transition metal oxide/reduced graphene oxide papers to produce flexible, free-standing and high areal mass hybrid paper electrode. The well separated reduced graphene oxide sheets decorated with transition metal oxide nanostructures have been observed, facilitating the access of electrolyte ions onto the high surface area of the hybrid paper electrode. This characteristic will enable the fabrication of thicker hybrid paper electrodes (up to  $150 \mu\text{m}$  thickness) and heavier areal mass (up to  $3.5 \text{ mg cm}^{-2}$ ), without sacrificing their mechanical integrity as well as gravimetric and areal capacitance. For instance, flexible and free-standing vanadium oxide/reduced graphene oxide paper electrode is able to achieve areal capacitance as high as  $1 \text{ F cm}^{-2}$ .

In addition, in order to elucidate the flexibility of hybrid paper electrode, supercapacitor device based on transition metal oxide/reduced graphene oxide hybrid paper electrode has been tested in its flat and bent state. The electrochemical performance of the supercapacitor device tested in its bent state remains similar to the one in its flat state, suggesting the good mechanical and electrochemical robustness of the device. Furthermore, supercapacitor device tested in its bent state also exhibits a good long term cycling stability, further supporting the