ABSTRACT

Title: "Study on the morphological effect of metal oxide based electrode materials for energy storage applications."
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As an intermediate energy storage device between dielectric capacitors and batteries, supercapacitors have attracted much attention due to their high power densities relative to secondary batteries and high energy density compared to traditional electric double-layer capacitors. Commercial supercapacitor utilizes high surface area carbon as an electrode to store charges. Unfortunately, these devices with carbon electrodes cannot demonstrate high energy density due to the physical storage mechanism. On the other hand, different transition metal oxides and few conducting polymer-based electrodes can store charges via fast and reversible faradic reaction with suitable electrolytes and demonstrate very high energy density. Ideal material for supercapacitor should have a high surface area, good electrical conductivity, optimum porosity, high specific capacitance, and excellent thermal and chemical stability. Apart from these parameters, morphology of the electrode material influences all these mentioned parameters and thus plays a vital role in determining the overall performance. An ordered morphology of the electrode material can enhance the surface area and can offer optimum porosity. A morphology where all the redox-active sites of the electrode material are electrically connected via some conductive pathways provides additional benefits of good conductivity and stability. In this thesis work, we have observed the effect of morphology on the electrochemical charge storage performance of the electrode material.

MnO_2 has been chosen for our first work as an electrode material. To stabilize against the volumetric strain during the repetitive charge discharge process, graphene oxide has been added to MnO_2 to form a composite. To study the effect of morphology, electrospinning technique has been used to fabricate fiber-like morphology of this composite. This morphology was chosen because of the fact that different redox-active sites can be electrically connected through the carbonized fiber structure. Detailed electrochemical performance testing showed that this ordered and well-connected morphology enhances the performance of this composite compared to the bulk sample having negligible interconnection between the redox-active sites. A high specific capacitance of ~893 F g^{-1} was obtained for this fibrous composite, whereas the bulk disordered sample offered 546 F g^{-1}. Using the fibrous sample as a positive electrode and activated carbon as a negative electrode, we fabricated an asymmetric two-electrode device which demonstrated a high specific capacitance of

212 F g^{-1} and a peak power density of 2.8 kW kg^{-1} and a peak energy density of 96.75 Wh kg^{-1}.

But electrospinning is a shallow yield process and is not suitable for mass production. To speed up the process of fiber production, we devised a simple Rotary Jet Spin (RJ-Spin) setup, which is identical to a candy floss machine. MnO_2/C fiber was fabricated using the RJ-Spin setup, and its electrochemical performances were evaluated to find the effect of fibrous morphology. A three-electrode-specific capacitance of 663 F g^{-1} was obtained for this sample. The two-electrode asymmetric device showed a specific capacitance of 136 F g^{-1}, a high power density of 3.2 kW kg-1, and a high energy density of 56.7 Wh kg^{-1}.

To reduce the resistance of MnO_2 based electrodes and enhance the charge storage performance, a bimetal oxide of Ni and Mn ($NiMn_2O_4$) was synthesized on nickel foam via a hydrothermal route. During the synthesis, different surfactants and mineralizer were used to tune the morphology of the synthesized $NiMn_2O_4$. By studying the detailed electrochemical performance, the effect of morphology of these samples was studied. It was seen that the addition of sodium dodecyl sulfate (SDS) as a surfactant resulted in an accumulation of spherical nanoparticles of $NiMn_2O_4$ with good interconnection between them. A specific capacitance of 1937 F g^{-1} was obtained for this sample coated on nickel foam. A two-electrode asymmetric device has been fabricated using the SDS-assisted hydrothermally synthesized powder sample as a positive electrode and activated carbon as a negative electrode. The device offers a specific capacitance of 270 F g^{-1} with excellent cyclic stability and good power and energy density.

As the conducting pathways provided by the nickel foam enhance the electrochemical performance, we tried to form $NiMn_2O_4$/Carbon fiber using the RJ-spin technique. For this purpose, the same SDS-assisted hydrothermally synthesized powder of $NiMn_2O_4$ was taken and was arranged in fiber form through RJ-spinning. Its electrochemical charge storage performance was compared to the powder sample to correlate the effect of morphology with the electrochemical performance. We have seen that the fibrous sample offered a specific capacitance of 460 F g^{-1} compared to the specific capacitance value of 405 F g^{-1} provided by the powder sample. It was also seen that the fibrous sample offered good rate capability and less resistance to current flow.

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