

Abstract/Synopsis: The main objective of this thesis is to develop polyaniline-based nanocomposite active electrode materials with high specific capacitance as well as cyclic stability, for supercapacitor applications. Intrinsic polymers play a key role in supercapacitor application because of their high corrosion resistance, flexibility, and good adhesion. Polyaniline is considered among all conducting polymers to be a promising supercapacitor material due to its superior properties such as multiple redox behavior, simple processability, high conductivity, and easy morphological alignment. Despite being one of the best supercapacitor materials, polyaniline has some disadvantages including poor cyclic stability (~1000 cycles) and low energy density. Several strategies have been adopted in the literature to overcome these issues. One of the strategies to develop nanocomposites of polyaniline with metal oxides and carbon-based materials. These nanocomposites show unique morphologies along with improvement in the specific capacitance and cycle life.

In this study, we have synthesized and evaluated the performance of several polyaniline-based nanocomposites. The first one we prepared was carbon-polyaniline nanocomposites namely, carbon nanotube-polyaniline, graphene oxide-polyaniline, and carbon nanotubes-graphene oxide-polyaniline. These nanocomposites benefit from the high specific surface area, high flexibility, and tensile strength of the carbon nanoforms, which improve the specific capacitance, allow high volumetric changes in the electrode material without degradation and as a result, provide better cyclic stability. The carbon nanoforms in polyaniline provide greater connectivity between the material domains. This enhanced connectivity and the resulting improved transport of charges could also be a key factor responsible for improving the cyclic stability of the polyaniline nanocomposite as a supercapacitor material. In any case, carbon-polyaniline nanocomposite supercapacitors show comparatively low specific capacitance (234, 265, 373F/g) values.

To solve this problem, a porous nickel cobaltite was incorporated into the polyaniline. Nickel cobaltite has high electrochemical activity and conductivity on account of the co-existence of two distinct cations in a single crystal. Porosity was introduced in nickel cobaltite by a glycine-assisted hydrothermal synthesis method. With the help of strong electrode/electrolyte interface, higher surface area ($176 \text{ m}^2/\text{g}$) because of the porous nature of glycine-assisted nickel cobaltite a moderate specific capacitance (688 F/g) with good cyclic stability (88% over 1000 cycles) was obtained. However, the specific capacitance has been moderately achieved, the cyclic stability will be lower if the cycles rise above 1000. Also, the electrochemical performance will be lost while making a two-electrode device due to poor adhesion.

To solve those problems polyaniline composite was made using carbon quantum dots which were hydrothermally synthesized using urea and glycine as a source. Polyaniline supplies pseudo-capacity for the composite and carbon quantum dots supplies a high surface area, enhanced electron transport, and potentially improved durability. When used as a precursor to polyaniline, CQDs are uniformly distributed, well-spaced, and isolated from each other without any agglomeration contributing to a long nanotube morphology. This type of long rod-like morphology leads to high interfacial interactions between polymers and CQDs, which can accommodate more ion transport and reaction sites, as well as the high specific capacitance of 871 F/g and 84.5 % retention in specific capacitance over 3000 cycles for a three-electrode system. The two-electrode device achieved a 161 mF/cm² areal-specific capacitance and cyclic stability of 85 % over 5000 cycles with this material.

The final material produced was a less time-consuming new organic nanostructure at a low-temperature simple heating method, which was later decorated with polyaniline. Because of the generation of more active charge storage sites leads to high interfacial interaction between organic nanostructures and polyaniline, this composite exhibits improved electrochemical performance in both liquid-based and solid-state systems. Excellent cyclic stability (85 % over 3000 cycles) and enhanced specific capacitance (779 F/g) have been achieved by a three-electrode system. The two-electrode device achieved an areal-specific capacitance of 650 mF/cm² with outstanding cyclic stability of 96 % up to 5000 cycles.

Although polyaniline has some superior properties, the low rate and cyclic stability are key issues associated with polyaniline electrodes. These issues were overcome by the efficient combination of polyaniline with the above-mentioned materials as composite electrodes.