Graphene and N-doped Graphene Coated with SnO₂ Nanoparticles as Super-capacitor Electrodes

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Super-capacitor is considered as a promising high-power source for digital communications and electric vehicles due to its high power capability and long cycle life compared with batteries. Carbon-based materials with high surface area, long cycle life and good mechanical properties are usually used in double-layer capacitors¹. As a novel carbon nanomaterial, graphene has attracted enormous interest due to its unique properties, such as extraordinary electrical properties and ultral-arge specific surface area. Recently, graphene sheets coated with SnO₂ nanoparticles have been synthesized to improve the capacity and cyclability of super-capacitors. It is known that defects and aggregation deteriorate electrical conducting property of graphene and influence the bonding between graphene and metal oxides. In this work, graphene- and nitrogen (N)-doped graphene-supported SnO₂ films were prepared and solid-state super-capacitors were fabricated employing graphene-based films and polyvinyl alcohol (PVA)-H₃PO₄ gel as electrodes and electrolyte, respectively. The electrochemical properties of the devices were studied.

Graphene and nitrogen-doped graphene were synthesized by a solvothermal method at 200 °C for 10-20 h, using tetrachloromethane (CCl₄, 2.0 mL), lithium nitride (Li₃N, 1.0 g), and potassium (K, 1.0g) as carbon precursor, nitrogen dopant, and catalyst, respectively. Graphene and N-doped graphene thin films were prepared by a blade coating method on glass slides, and then subjected to a heat treatment at 250°C for 1h to remove organic materials. SnO₂ nanoparticles supported on graphene films were synthesized through a chemical-solution routine using $SnCl_2$ as a precursor ². 100 mg of SnCl₂ was dispersed in 10mL of deionized water, and 175 μL of hydrochloric acid (HCl, 38%) was added to the mixture. The solution was then sonicated for 10 min. After sonication, the graphene film was immersed in the solution for 30 min-60 min, and rinsed with deionized water followed by drying in air.

Graphene based super-capacitors were fabricated using polyvinyl alcohol (PVA)-H₃PO₄ gel (0.8 g H₃PO₄ in 10 mL 10% PVA solution) as electrolyte, which was placed between two pieces of graphene based electrodes. The as-fabricated devices were dried at room temperature atmosphere, and subjected to electrochemical in measurements for the evaluation of their super-capacitor behavior. Scanning electron microscopy (SEM) and transmission electron microscope (TEM) were used to characterize the morphology and structure of the obtained graphene-based materials. Energy dispersive X-ray spectrometer (EDS) and X-ray diffraction (XRD) analyses were carried out to investigate the composition and crystal structures of SnO₂/graphene. The electrochemical (EC) performance of as-fabricated super-capacitors was measured using cyclic voltammetry (CV), cyclic charge/discharge (CCD), and electrochemical impedance spectroscopy (EIS).

For instance, as given in Figure 1, it was observed that the incorporation of SnO_2 significantly enhanced the electrochemical performance of graphene based super-capacitor (Figure 1a), and the CV curve of SnO_2 /graphene super-capacitor displayed nearly rectangular shape, indicating that an efficient electrical double layer was established at both graphene based electrodes (Figure 1b).



Figure 1 The performance of graphene-based supercapacitors: (a) Cyclic voltammetry curves obtained at 100mV/s for graphene and SnO_2 /graphene, and (b) cyclic voltammetry curves obtained at different scan rates for SnO_2 / graphene capacitor.

In summary, graphene and N-doped graphene with few defects were prepared by a solvothermal method. Electrochemical measurements indicate that the incorporation of SnO_2 nanoparticles can enhance the performance of graphene-based super-capacitors significantly.

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