Abstract

Supercapacitors, with their high power density, high specific capacitance, and faster charge-discharge rate have attracted great attention for their high stability and long-term use as energy storage devices that complement batteries. The performance of a supercapacitor depends on various factors, including the nature of the electrode material and electrolytes. In this dissertation, two-dimensional (2D) materials have been studied as supercapacitor electrode materials due to their exceptionally high surface area, active sites, and flexibility. Herein, 2D materials such as graphene oxide (GO), GO-TiO_2 nanocomposites, WS_2 and siloxene are synthesized on large scale with a simple and solution processed method. Morphology, crystal structure, thickness, elemental composition, and surface area of the synthesized 2D materials are investigated through various characterization techniques such as scanning electron microscopy (SEM), transmission electron microscopy (TEM), x-ray diffraction, Raman, UV-absorption, x-ray photoelectron spectroscopy, BET, Energy dispersive spectroscopy, etc. We have developed an easy, fast, scalable one-pot controlled sol-gel method to synthesize GO-TiO_2 nanocomposite of different compositions. We have shown that the optimum composition of GO-TiO_2 nanocomposite (TG25) exhibits a high areal specific capacitance of 73.43mF/cm^2 at a current density of 0.5 mA /cm^2 with good flexibility and excellent cycling stability. We have fabricated devices using binder-free siloxene nanosheets with various electrolytes to get a deep insight into the importance of an appropriate combination of electrode-electrolyte systems, proton coupled electron transfer (PCET) kinetics, and the charge storage mechanism to enhance the performance of the devices. The novel combination of binder-free siloxene electrode and PVA/H_3PO_4 gel electrolyte provides a high specific capacitance of 33.1 F/g (~ 99.31 mF/cm^2) at 10mV/s with ~ 99% capacitance retention over 10,000 cycles. Next, we have exfoliated WS_2 using different intercalating agents such as ethylenediamine and lithium iodide. We have synthesized 2D WS_2 with high yield by a simple LiI assisted modified liquid exfoliation method. The applicability of WS_2 as a supercapacitor electrode material is studied through Kelvin probe force microscopy and XRD analysis. Finally, sustainable and environmentally friendly supercapacitor devices are fabricated using ionic liquid (IL) salt and polyvinyl alcohol composite as gel electrolyte with WS_2 nanosheets. The effect of conductivity and viscosity of ionic liquid based polymer gel electrolytes with varying molar concentrations of IL on the WS_2 electrode based supercapacitor performance is investigated.

Keywords: Supercapacitor, EDLC, pseudocapacitor, hybrid supercapacitor, solid-state supercapacitor, two-dimensional material, Graphene Oxide, Nanocomposites, WS_2, Siloxene, Ionic liquid, PCET, hybrid charge storage mechanism