Index No. 11/18/Phys./25

ABSTRACT OF THE DISSERTATION

Experimental investigation of the ionic conduction and dielectric properties of chitosan based solid biopolymer blend and its nanocomposites for solid-state supercapacitors : EDLC to Hybrid

The increasing demands for long-lasting and lightweight portable electronic devices suggest intense research for supercapacitors having higher power density than batteries and conventional capacitors, with solid state electrolytes. Natural polymers such as chitosan has attracted huge attention as solid state electrolytes since the last few decades owing to their low cost, easy availability from shells of shrimps and crabs, principal bio-degradation properties and their ease of fabrication as thin films which are mechanically very strong, while spinel ferrites, $\boldsymbol{M}Fe_2O_4$ or $\boldsymbol{M\acute{M}}FeO_4$ (where $\boldsymbol{M}$ or $\boldsymbol{\acute{M}}$= Mn, Ni, Zn, Co, etc.) are being considered as fascinating electrode materials in supercapacitor applications for the last several years due to their ability to exhibit different redox states, impressive electrical properties and good electrochemical stability. The objective of this dissertation is to develop a high-performance solid-state supercapacitor by enhancing the ionic conductivity and dielectric properties of an environmental friendly and cost-effective solid polymer electrolyte which involves both faradaic and non-faradaic energy storage mechanisms at the electrode, to reach the increasing power demands in our society. A solid polymer electrolyte (SPE) intended to use in solid-state supercapacitors must have sufficiently high ionic conductivity. Ionic conductivity of the solid electrolytes depends upon the charge carrier concentration which can be changed by doping the biopolymer (CS) with an ionic salt ($LiClO_4$). Firstly, the ionic conductivity of pure chitosan-lithium perchlorate SPE has been investigated which is found to be of the order ~ 10^{-5} S/cm. But the conductivity value is still very low for its implementation in supercapacitor applications. Plasticization and blending with other polymers are done in order to improve the ionic conductivity of the solid electrolytes further. Blending of CS with natural polymers such as PS and CMC improves the conductivity of the solid electrolytes upto an order ~ 10^{-4} S/cm, which is still quite low. Our study reveals that blending CS with PVDF (a synthetic polymer) exhibits the highest conductivity of the solid electrolyte which is of the order ~ 10^{-2} S/cm, whereas blending with another synthetic polymer (PMMA) gives the ionic conductivity value of ~ 10^{-3} S/cm. Dielectric properties of the blended polymers has been tailored by grafting with graphene oxide (GO) and multi-walled carbon nanotube (MWCNT). A significant enhancement of dielectric constant with reduced loss factor is achieved due to the nanocomposite formation. The relaxation function within the solid-state electrolytes is found to be highly non-exponential, suggesting non-Debye relaxation. The intra ion-diffusion processes has been entirely analyzed in the light of anomalous fractional diffusion model which reveals dominating anomalous diffusion behavior of the ions inside the electrolytes. On the application front, a coin type unique symmetric supercapacitor has been developed respectively with active carbon, $MnCoFeO_4$ and rGO/$MnCoFeO_4$@Ag nanocomposites as electrode materials. We have discussed the poor accessibility of the solid-state electrolyte ions to the active sites in the nanoporous electrode materials unlike liquid electrolytes, thus demanding the evaluation of inter ionic transport properties of the charge carriers in order to determine the effective specific capacitance of the electrode materials. It has been observed that the effective specific capacitance is increased significantly with $MnCoFeO_4$ compared to that of active carbon. On decorating $MnCoFeO_4$ with silver (Ag) nanoparticles and anchoring them onto graphene sheets enhances the effective specific capacitance remarkably. Our study reveals an enormous enhancement of effective specific capacitance ~ 750 Fg^{-1} which makes our system a promising candidate for future development of safe and cost effective electrochemical hybrid solid state supercapacitors.

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