Abstract

With the advancement of society, net energy consumption is increasing exponentially. Although the inception of nuclear and renewable energy reduces the proportion of use of fossil fuels by around 15%, still consumption of fossil fuels has increased many folds in the last hundred years. The increased use of fossil fuels is solely responsible for the emission of CO_2 and other greenhouse gases. Thus, widespread use of renewable energy sources is essential to curb the emission of pollutant gases. However, most of the renewable energy harvesters (e.g. solar and wind) generate electrical energy with irregulated consistency. So, smart grids and power efficient energy storage systems (ESS) are necessary for smooth regulations of harvested energy from renewable sources. Alongside this, transportation sector is responsible for enormous emissions of air pollutants. Particularly for on-road transportation, electric vehicles could restrict the emissions. Although hydrogen fuel cells and batteries can carry a gigantic amount of energy (in small device volume), they doesn't match the combustion engine's efficiency in terms of their power density. To improve the accelerating and decelerating of electric vehicles, it is necessary to develop a hybrid energy storage system by coupling high power density supercapacitors with high energy density batteries or fuel cells. Supercapacitors have become essential for ESS and other high power applications due to their high power density, adequate energy density, and long lifetime. Performance of the supercapacitors mostly depends on their energy storage mechanism and device architectures. In terms of charge storing mechanism, supercapacitors can be classified as; (i) electric double layer capacitors (EDLC) and (ii) pseudocapacitors. EDLC stores charge electrostatically without involving in any faradaic reactions, which enables much faster charge transfer and ultrahigh lifetime for EDLCs than the pseudocapacitors. On the contrary, pseudocapacitors stores charge by involving surface limited, highly reversible faradaic processes. Due to this, pseudocapacitors' energy density is higher than EDLCs with a sacrifice in charge transfer rate and lifetime. An efficient supercapacitor can be developed either by increasing the energy density of EDLCs or increasing the power density of pseudocapacitors along with the improved compactness of the device architecture.

This thesis demonstrates various methodologies to increase energy and power density (and related specific capacitance) of supercapacitors by (i) defect induced 3D RGO forest structure, (ii) alkali metal incorporated biomass derived carbon (BDC) electrodes, (iii) development of V_2O_5 based power efficient extrinsic pseudocapacitors and (iv) fabrication of improved in-plane screen-printed micro supercapacitor (MSC). Here the net measurable, specific capacitance of the RGO forest structure was improved by increasing the minimal quantum capacitance (C_Q), usually in series with the EDL capacitance (C_{dl}) of RGO by tuning the defects in RGO flakes through uv-ozone treatment. The specific capacitance of alkali metal incorporated biomass derived carbon electrode was improved by the tunable porosity and specific surface area of the electrode materials achieved through different activating agents and carbonization temperature. Also, the incorporated alkali metals in BDC takes part in some parasitic electrochemical process, which increases their coulombic efficiency beyond 100%. An asymmetric supercapacitor was fabricated with the optimized BDC material as the negative and V_2O_5 micro flakes as the positive electrode with the presence of PVA@KOH hydrogel as an electrolyte. Generally, V_2O_5 acts as a battery electrode in lithium electrolytes. Here V_2O_5 has been transformed into an extrinsic Li-ion pseudocapacitor with increased power density by structurally modifying V_2O_5 which changes the diffusion lengths for Li-ions. Finally, a set of compact in-plane micro supercapacitors (MSCs) were fabricated using layer-by-layer screen printing of silver current collector and BDC ink (screen printable). Here, reduction in equivalent series resistance (ESR) and improvement in lifetime of the MSCs were achieved by fully wrapping the current collector tracks by the electrode material. Specifically, a power density of 656.25 mWcm^{-3} for the RGO forest electrode based (aqueous electrolyte) sandwich device is increased to 18000 mWcm^{-3} in the case of ionic liquid based ($EMIMBF_4$) in-plane MSC with wrapped current collector architecture, along with almost thousand fold increase in volumetric energy density.

Keywords: Supercapacitors, extrinsic pseudocapacitors, uv-ozone treatment, 3D RGO forest, biomass derived carbon, in-plane micro supercapacitor, screen printing, structural modification of V_2O_5.