

## Abstract

Increasing requirements of multifunctional electronic gadgets have attracted substantial research curiosity particularly in the arenas of maintenance-free portable energy harvesting and storage systems. Essentially, piezoelectric/triboelectric-based nanogenerators, and photovoltaic cells are widely exploited as ambient energy harvesters to harvest numerous forms of available energies while batteries and electrochemical supercapacitors are expansively exploited to store electrochemical energy for sustainable energy/power generations. Intriguingly, supercapacitors have enticed modern scientific communities/industries owing to their several inherent features like; higher power density, elevated specific capacitance, decent cyclic stability, lesser toxicity, inferior heat developments during recurrent charging, and fast charge-discharge rates which are exceedingly beneficial for consumer electronics. In facts, electrochemical supercapacitors form a bridge in-between the conventional dielectric-type capacitors (with substantial power density and lower energy density) and traditional batteries (with substantial energy density and inferior power density). The prime components of a supercapacitor cell are its electrodes, electroactive materials, separator and electrolyte. According to device configurations, supercapacitors can be classified into two different kinds viz. symmetric-type supercapacitors (SSCs) and asymmetric-type supercapacitors (ASCs). Amongst them, ASCs show greater specific capacitances, energy, power densities and wide working voltage windows than SSCs. Further, inclusion of moderate-to-high degrees of flexibility/stretchability within the ASCs will be extremely advantageous for wearable energy storage application. According to the electroactive materials side, four-distinct capacitive materials are widely utilized in SSCs/ASCs such as; electric double layer capacitive-types (EDLC), pseudocapacitive (PC), battery-types and their hybrid composites. Furthermore, to alleviate the concerns like restricted operational times and frequent charging of SSCs/ASCs, scientists have recently developed various piezoelectric/triboelectric nanogenerator driven self-charging supercapacitors for maintenance-free operations of personalized electronic gadgets.

In this thesis, complete endeavours have been dedicated to design/fabricate/characterize miscellaneous ASCs and flexible self-charging ASCs (piezoelectric/triboelectric nanogenerator driven) by employing Faradaic-type transition metal oxides/hydroxides/their hybrid composites (as positive electroactive materials) and high surface area graphene-based nanoarchitectures (as negative electroactive materials) in presence of KOH-based liquid/solid-state electrolytes. The structural, morphological, and electrochemical characteristics of all the synthesized component electroactive materials have been analysed and illustrated comprehensively in this thesis. Some prototypes of self-charging ASCs were developed either by using bio-piezoelectric separator or triboelectric nanogenerators. Typical self-charging mechanisms (instant energy harvesting with consequent storage) of such self-powered systems under applied external stresses/deformations and their performances along with potential application arenas are also highlighted succinctly.

**KEYWORDS:** Energy harvesters; Supercapacitors; Power density; Electroactive materials; Piezoelectric; Triboelectric; Self-charging.