Supercapacitors, the devices that connect the gap between batteries and conventional capacitors, have recently attracted significant attention due to their high specific capacitance, substantially enhanced power and energy densities, and extraordinary cycle life. In order to realize even better performance with supercapacitors, rejuvenated effort towards developing nanostructured electrodes is necessary.

In this dissertation, several strategic directions of nanoarchitecturing the electrodes to enhance the performance of supercapacitors are investigated. An introduction and background of supercapacitors, which includes motivation, classification and working principles, recent nanostructured electrode materials studies, and devices fabrication, are initially presented. A facile method, called Spin-on Nanoprinting (SNAP), to fabricate highly ordered manganese dioxide (MnO2) nanopillars is introduced. The SNAP method further modified to develop carbon nanoarray electrodes is also discussed. Subsequently, a template-free method to develop high aspect ratio copper oxide nanowhiskers on copper substrate is presented, which boosts the surface area by 1000 times compared to non-nanostructured copper substrate. Electrochemically deposited MnO2 on the nanostructured substrate provided a specific capacitance of about 1379 F g\(^{-1}\), which is very close to the theoretical value (~ 1400 F g\(^{-1}\)) due to this efficient nanostructure design. In addition, a novel method to decorate metal nanoparticles on graphene aerogel, which considerably enhances the electronic conductivity and the corresponding specific capacitance, is demonstrated. Moreover, ferric oxide (Fe2O3) nanorods prepared by a simple hydrothermal method is discussed. Asymmetric devices assembled based on Fe2O3 nanorods and MnO2 nanowhiskers show excellent electrochemical properties. The devices not only display the capability to store energy but also transmit electricity through the inner copper core. These two functions are independent and do not interfere with each other. Finally, a summary of this dissertation as well as some potential future directions are presented.