Organic and perovskite solar cells have recently attracted significant attention due to its flexibility, ease of fabrication and excellent performance. In order to realize even better performance for organic and perovskite solar cells, rejuvenated effort towards developing nanostructured electrodes and high quality active layer is necessary. In this dissertation, several strategic directions of enhancing the performance of organic and perovskite solar cells are investigated. An introduction and background of organic and perovskite solar cells, which includes motivation, classification and working principles, nanostructured electrode materials and solvent effect on active materials, and devices fabrication, are presented. A facile method, called Spin-on Nanoprinting (SNAP), to fabricate highly ordered ZnO-AgNW-ZnO electrode is introduced to enhance the performance of organic solar cell. Subsequently, a ternary solvent method is developed to fabricate high Voc PTB7:ICBA solar cells. The performance of the devices improved about 20% compared to those made by binary solvent method. In order to understand the fundamental properties of the materials ruling the performance of the PSCs tested, AFM-based nanoscale characterization techniques including Pulsed-Force-Mode AFM (PFM-AFM) and Mode-Synthesizing AFM (MSAFM) are introduced. These methods are used to study the morphology and physical properties of the structures constitutive of the active layers of the PSCs. Conductive-AFM (cAFM) studies reveal local variations in conductivity in the donor and acceptor phases as well as an increase in photocurrent measured in the PTB7:ICBA sample obtained with the ternary solvent processing technique. Moreover, efficient perovskite solar cells with good transparency in the visible wavelength range have been developed by a facile and low-temperature PCBM-assisted perovskite growth method. This method results in the formation of perovskite-PCBM hybrid material at the grain boundaries which is observed by EELS mapping and confirmed by steady-state photoluminescence (PL) spectra and transient photocurrent (TP) measurements. This method involves fewer steps and therefore is less expensive and time consuming than other reported methods. In addition, we report an all solid state, energy harvesting and storing (ENHANS) filament which integrates perovskite solar cell (PSC) on top of a symmetric supercapacitor (SSC) via a copper filament which works as a shared electrode for direct charge transfer. Developing ENHANS on a copper filament provides a low-cost solution for flexible self-sufficient energy systems for wearables and other portable devices. Finally, a summary of this dissertation as well as some potential future directions are presented.