Abstract

The global climate change is calling for an increasing implementation of new energy sources to sustain fuel-based technologies and processes. Simultaneously, the society is moving at an impressive pace towards an ever-increasing integration of devices within the objects of our everyday lives. This vision can be realized through the development of photocapacitors - bifunctional devices that combine solar harvesting and energy storage within compact units capable of off-grid charging and long-term stability. The highest level of integration is provided by material systems that can perform both functionalities at the same time. The materials system studied within this thesis is based on metal oxide (MO) nanocrystals (NCs), cost-effective compounds with tuneable optical and electrical properties, arising from doping with heteroatoms or defects, or post-synthetic approaches, like photodoping. In the photodoping process, photocharges are created when illuminated with light beyond their bandgap. In proper conditions these charges are stored within the NC and can be extracted for their later use. This process was observed in several MO NC systems, such as indium tin oxide or doped zinc oxide. However, so far it has remained a solution-based process, still not efficiently translated into solid state technology. Within my thesis, I focused on the development of bifunctional devices from MO NCs, capable of harvesting light and storing the charges in two-terminal devices. I have focused on tungsten oxide (WO_3) NCs, as a widely used materials system with excellent charge retention capability and optimal bandgap. In the work herein presented, I investigated different synthetic routes to obtain control on substoichiometric WO_3 ($WO_{2.69}$) NCs dimensions, accounting for the optoelectronic properties related to morphology. Furthermore, I focused on the processing of the material into thin films for the fabrication of photoactive layers. I investigated the mechanism of the photodoping on the NC solution, in view of translating the photostorage properties of $WO_{2.69}$ in solid state. Finally, I proceeded with the fabrication of 2-terminal devices, whose performances were examined with a thorough photoelectrochemical characterization. I also tested different designs and combinations of materials with the goal of studying the best operative conditions and the most promising architecture for the devices. With this thesis, I thus laid the foundations for the investigation of WO_3 as bifunctional material, and for a renewed focus on 2-terminal photocapacitors as valid medium for easily and effectively combining light harvesting and energy storage.