

Abstract

Understanding the electrical properties governing systems for energy conversion and storage is essential for improving device efficiency and longevity. This thesis investigates organic solar cells and hydrogen production systems through photoelectrochemical methods, with a particular focus on the development and characterization of organic-based active layers for solar cells and as photoelectrodes. Alternating potential techniques, including impedance spectroscopy (IS) and electrochemical impedance spectroscopy (EIS), are employed to analyze both the devices and their active layers, providing insights into their underlying dynamics.

Impedance spectroscopy and electrochemical impedance spectroscopy emerge as indispensable tools in unraveling the electrical behavior of these systems. These techniques offer a non-destructive means to probe the electrical response of materials and devices over a broad frequency range, providing valuable insights into their electrical properties and underlying mechanisms. By analyzing the impedance spectra, key parameters such as charge carrier mobilities, recombination rates, and interface properties can be extracted, offering crucial information for optimizing device performance.

Beyond performance analysis, impedance techniques also facilitate the determination of critical thermodynamic factors and kinetics governing energy conversion processes. Through impedance spectroscopy, we can elucidate thermodynamic parameters such as density of states, band positions, builtin potentials, and density of defects within the materials. Additionally, these techniques enable the determination of kinetic parameters such as exciton lifetimes, contributing to a deeper understanding on charge carrier dynamics and recombination processes in these systems.

This thesis includes the development of experimental setups and methodologies. A photoelectrochemical setup was constructed to meet the specific requirements of this research. Furthermore, novel characterization techniques were developed, and the synthesis of organic nanoparticles was implemented in the laboratory for the first time. This work also integrates impedance techniques with photoelectrochemical methodologies, broadening the capabilities of the laboratory for future research.

Chapter 1 provides a brief background on organic semiconductors, solar cells, and photoelectrochemical water splitting, along with an overview of the theoretical principles behind impedance spectroscopy. It also presents the state of the art in solar cell and photoelectrochemical system research, as well as the application of IS in these fields. Schemes and images of the experimental setups are included at the end of the chapter. Chapter 2 presents an impedance analysis of organic solar cells with a nanoparticle-based active layer, comparing their electrical behavior to traditional bulk heterojunction solar cells. It also introduces the use of Energy Resolved-Electrochemical Impedance Spectroscopy (ER-EIS) to study the impact of film morphology on the electronic structure of the active layer components. Chapter 3 explores the characterization of donor:acceptor nanoparticles through photoelectrochemistry, leading to the development of a novel photoelectrode for hydrogen evolution. Chapter 4 focuses on the development and study of an organic layerbased photoanode with a polydopamine protective layer, characterized using photoelectrochemical methods.

Chapter 5 presents the use of IS for characterizing organic solar cells and is divided into two sections. The first section examines an organic solar cell incorporating an ascorbic acid interlayer as a protective component, with IS employed to study the impact of this layer on the

photodegradation of the device. The second section applies ER-EIS to a thermal and photodegradation study, providing insights into defect formation within the band gap.

Chapters 2 and 4 contain manuscripts prepared for submission. These were modified to incorporate supplementary material into the main text, and figure numbering was adjusted for improved readability.

Chapter 5 presents results from contributions to two published papers in which I am a co-author.

Each manuscript contributes to the broader understanding of these energy systems, offering insights into their electrochemical and electrical characteristics relevant to advanced energy conversion and storage. The findings presented in this thesis provide a foundation for optimizing organic-based energy conversion and storage devices through impedance spectroscopy and electrochemical impedance spectroscopy, supporting the development of more efficient and durable energy technologies.