Center for Soft PhotoElectroChemical Systems (SPECS)
EFRC Director: Erin Ratcliff
Lead Institution: University of Arizona
Class: 2022 – 2026

***Mission Statement**: SPECS will understand the factors controlling charge and matter transport processes in inexpensive, scalable, and durable π-conjugated polymer (plastic) materials. We will explore the factors across spatiotemporal scales that underpin emerging energy conversion technologies to influence the formation of fuels, such as H_2, from sunlight and develop new approaches to energy storage.*

The diverse and engaged SPECS team includes synthetic, computational, and experimental experts working across three interconnected thrusts (**Figure 1**) involving π-conjugated, polymeric (plastic) semiconductor materials: **Hybrid Electrical-Ionic Charge Transport** (Thrust 1), **Charge Transfer and Energy Cascades** (Thrust 2), and **Durability** (Thrust 3). SPECS will fill crucial knowledge gaps that limit the introduction of these materials into the U.S. energy portfolio including: i) understanding and control of interface molecular composition and structures dictating charge (electrical and ionic) transport, ubiquitous across all electrochemical energy conversion and energy storage applications; ii) understanding dynamic responses of interfaces and mechanisms that drive (photo)-electrochemical charge transfer processes; and iii) creation of design guidelines that enable robust π-conjugated polymeric energy conversion and storage materials.

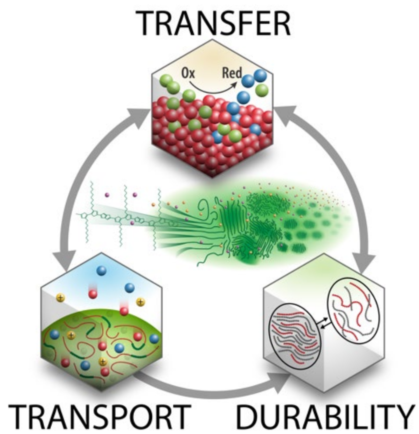

Figure 1. SPECS thrusts working synergistically to understand charge and matter transport processes in soft materials.

Direct outcomes of SPECS will be mechanistic insights and new physiochemical models of fundamental structure-property relationships of π-conjugated polymer platforms. Models based on fundamental chemistry will drive the search for anode and cathode materials at electrochemically reactive interfaces (catalysis) and buried atomic-to-nano interfaces that comprise the bulk (storage). These mechanistic insights require developing new measurement science approaches, which will be a high impact legacy of SPECS. We will advance characterization and operando approaches, under both environmental and operational "stresses," examining materials in near-surface, interface, and interphase regions, under conditions that push soft materials and platforms far from equilibrium. Fundamental insights gained by SPECS will translate across *Science for Clean Energy* applications, including connections to existing BES (EFRC and Hub) programs. SPECS research products will be disseminated to the greater scientific community through high-impact journal articles and creation of new intellectual property. SPECS represents a scientifically and demographically Diverse and Inclusive Team that fosters new knowledge discovery coupled to unique mentoring and student training, targeted to development of an inclusive next generation workforce needed to meet U.S. 2050 clean energy goals as well as Energy Justice initiatives.

To achieve SPECS' goals, we are organized into three synergistic and interconnected research thrusts. Thrust 1 focuses on comprehensive characterization of the complex π-conjugated polymer/electrolyte structures that control ion and polaron transport and energy storage processes. We will emphasize understanding fundamental design principles that drive the dynamic response (picosecond-to-second) of these systems at atomic-to-micron length scales. Thrust 1 includes a tight feedback loop among theory/modeling, materials design, synthesis, processing, and characterization to link complex electronic, electrochemical, and physical phenomena. Objectives include: 1.1) describe the physical structure of polymer-electrolyte interphases and how dynamic versus static structural disorder affects relevant transport processes, 1.2) define structure-property relationships controlling hybrid electrical-ionic transport and rates of charge/discharge in polymer-electrolyte systems, and 1.3) quantify strengths of ion-polaron interactions and describe how these interactions impact hybrid electrical-ionic transport.

Thrust 2 will understand and design π-conjugated polymer photocathodes that are tailored for efficient, directional electron and hole transfers as well as long-lived charge separation needed to drive charge (energy) for fuel-forming reactions, such as H_2 evolution. Data from theory/modelling, spectroscopy, and structural characterization will be critical to elucidate mechanisms of energy and charge transfer that we will leverage to benchmark and improve performance in relevant electrochemical π-conjugated polymer platforms. Within Thrust 2, SPECS aims to tailor interactions of complex phenomena to achieve integrated multicomponent systems. This thrust is tightly linked to efforts of Thrust 1 and ultimately Thrust 3 to ensure new durable energy conversion materials. Objectives include: 2.1) define the equivalent of an electric double layer in increasing complex π-conjugated polymer/electrolyte interfaces under operando (applied bias and/or light), 2.2) describe mechanisms of electron transfer in outer sphere redox reactions, and 2.3) investigate photoelectrochemical mechanisms — leading to solar fuel formation (H_2).

Thrust 3 will establish molecular scale understandings of π-conjugated polymer degradation and its mitigation, leading to electrochemical/photoelectrochemical systems that simultaneously exhibit high performance and long lifetimes. Durability of these materials is determined by a combination of bond forming/bond breaking, microstructure, and long-range connectivity of mechanical, thermal, optical, electronic, and electrochemical properties. Thrust 3 objectives include: 3.1) determine the chemical degradation pathways and eliminate detrimental chemistries, 3.2) evaluate long-time, operando-induced morphological changes and mechanical failures providing design guidelines to improve durability, and 3.3) manipulate local chemical and structural environments to increase durability. These three thrusts working cooperatively together will allow SPECS to gain a fundamental understanding of low-cost, durable energy conversion and energy storage technologies leveraging soft materials.

SPECS	
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