

DYNAMICS IN A BROAD SCALE

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Understanding and utilizing dynamics, at various time scales and system sizes, represent an underlying theme for most research projects in my group. Subpicosecond charge separation (at a molecular scale) and millisecond to second fluid dynamics (in microfluidic devices, at micrometer and millimeter scales) represent some of the extreme length and time scales that we investigate. A range of synthetic, analytical, design and fabrication techniques, broadly used in my lab, offer a liaison between basic science and applied engineering.

In relevance to solar energy conversion, the first part of the talk will focus on the kinetics of photoinduced charge transfer mediated by biomimetic and bioinspired systems. Protein-mediated electron-transfer processes sustain a broad range of redox functions in biological systems, such as respiration and photosynthesis. Therefore, such biological systems are indispensable “working” models for molecular design and for the development of materials for solar-energy-conversion and electronics applications. Examples will illustrate the role of solvent polarity, media viscosity and triplet formation for generating long-lived charge-separated states in biomimetic systems. Modulation of charge transfer by local electric fields, however, presents an alternative approach for achieving long-lived charge-separated states. Macromolecular electrets provide local field gradients in the order of 0.1 GV/m and degeneracy of charge-transfer states essential for directionality of electron and hole entrainment. (Electrets are the electrostatic equivalent of magnets: i.e., they possess ordered electric dipoles.) The presentation will cover our designs of bioinspired electrets and discuss their properties.

The second part of the talk will cover our advances in single-molecule force-modulated kinetics. This methodology offers unique capability for direct observation of a range of structure-function relations. For example, it is the only experimental approach that allows direct estimation of the displacements of the transition states during bimolecular interactions. Despite the conceptual simplicity of single-molecule force techniques, certain experimental design challenges, which will be discussed, have prevented such methodologies from becoming routinely used tools. Employing controlled surface chemistry, for example, allows us to address some of these issues.