Nanoscale Platform Embedded Reactions for Enhanced Chemical Transformations

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The overarching goal of our program is to establish intelligent control over molecular transformations at small length and time scales – enabling the construction of materials and structures that can sense molecular characteristics, *e.g.* size, charge, molecular shape, *etc*. and generate control signals that effect downstream molecular processing based on those characteristics. Specifically, these experiments seek to manipulate, (separate, isolate, react, detect) low-mass samples with the same precision and level of control currently possible with bench-scale samples by combining microfluidic and nanofluidic structures with advanced methods for controlled molecular assembly and guided by state-of-the-art simulations. The strategies used here target nanostructures in which fluids are moved by electrokinetic (EK) flow and which utilize electrochemical reactions for sensing and/or treatment. In order to fully realize the promise of this approach, the capacity to carry out controlled electron transfer reactions in the presence of EK flow in nanoscale architectures is being developed using a geometry called the embedded annular nanoband electrode. The fundamental chemical and physical factors that determine how electrified fluids couple to working electrodes at the nanoscale are being elucidated, and the spatial distribution of potential under EK flow in hybrid microfluidic-nanofluidic architectures is being studied using both experiments and simulations. We are also exploring the effect of electrode configuration (geometry) and placement, where the working electrode is floating and where it is referenced to a quasi counter/reference electrode. In addition, model reactions, especially those linked to pH changes that can be sensed optically, are being used to characterize the current efficiency of nanochannel-confined electrochemical reactions. Supporting all three of these objectives, plasmonic sensing strategies are being developed that are efficiently matched to nanoscale architectures. In particular, plasmonic sensing in extraordinary optical transmission structures is especially well-matched to the coupled EK flow-electrochemical approaches being developed. This work is supported by the National Science Foundation through the Center for Advanced Materials for the Purification of Water with Systems and by the Department of Energy Basic Energy Sciences.