Redox Mediators Coupled to Surfaces and Supports: Operation of Enzymatic Biofuel Cells in Physiological Buffers, Human Saliva and Blood

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An overview is presented of the strategies that we have adopted towards the integration of enzymes and redox mediators capable of efficiently transferring electrons between enzymes and electrodes; focusing on the ambitious goal of developing an implantable (or portable), miniature, membrane-less enzymatic fuel cell (EFC) by exploiting enzymatic oxidation of fuels, such as glucose, coupled to the enzymatic reduction of dissolved dioxygen [1-11].

One approach focuses on synthesis of a variety of osmium-based redox mediators for incorporation into enzyme electrodes, with screening of mediator selection based on choice of enzyme and/or immobilization strategy. We have synthesized a library of osmium-based redox mediator complexes and polymers possessing a range of redox potentials and various functionalized ligands. These redox mediators can be subsequently incorporated into different polymeric supports or tethered to different surfaces utilizing different coupling or ligand-exchange chemistries. Addition of nanostructured supports within enzyme electrode films are shown to have a beneficial effect with an increase in loading and retention of redox mediators and enzymes observed, leading to improvement to current output and signal stability.

In this talk we will discuss co-immobilization of separate multi blue copper oxidases, a Myceliophthora thermophile laccase, Streptomyces coelicolor laccase and a Myrothecium verrucaria bilirubin oxidase with osmium redox polymers or complexes possessing an amine-terminated molecular tether can produce current for oxygen reduction in the presence of multi-walled carbon nanotubes on graphite electrodes under pseudo-physiological conditions. We will also present a comparison of the glucose oxidation response of films prepared varying the sugar-oxidizing enzyme catalyst, in an attempt to provide for even greater current production at the anode, for application to EFCs. Modifying the redox enzymes through de-glycosylation or mutation provides another route for optimization of their properties for application to fuel cell catalysis.

Finally, we report on the operation of assembled, membrane-less, enzymatic fuel cells in physiological solutions, un-stimulated human saliva and blood; providing power to enable wireless transmission of sensing data.

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